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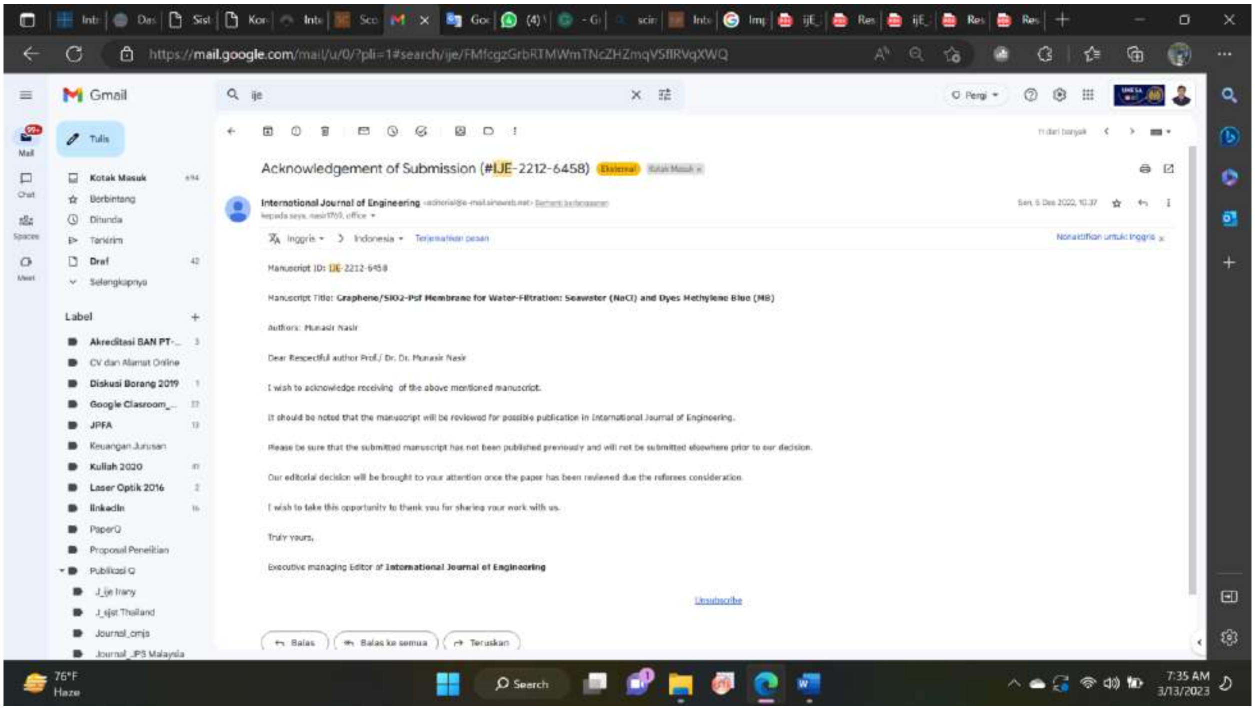

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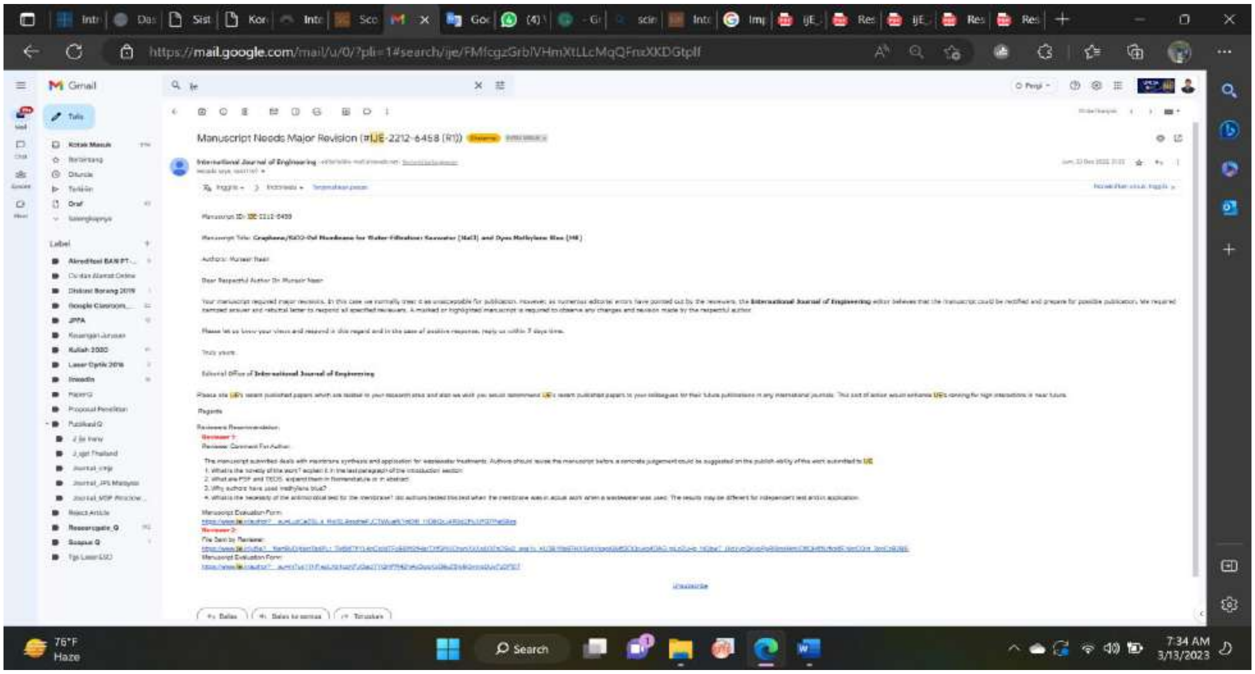

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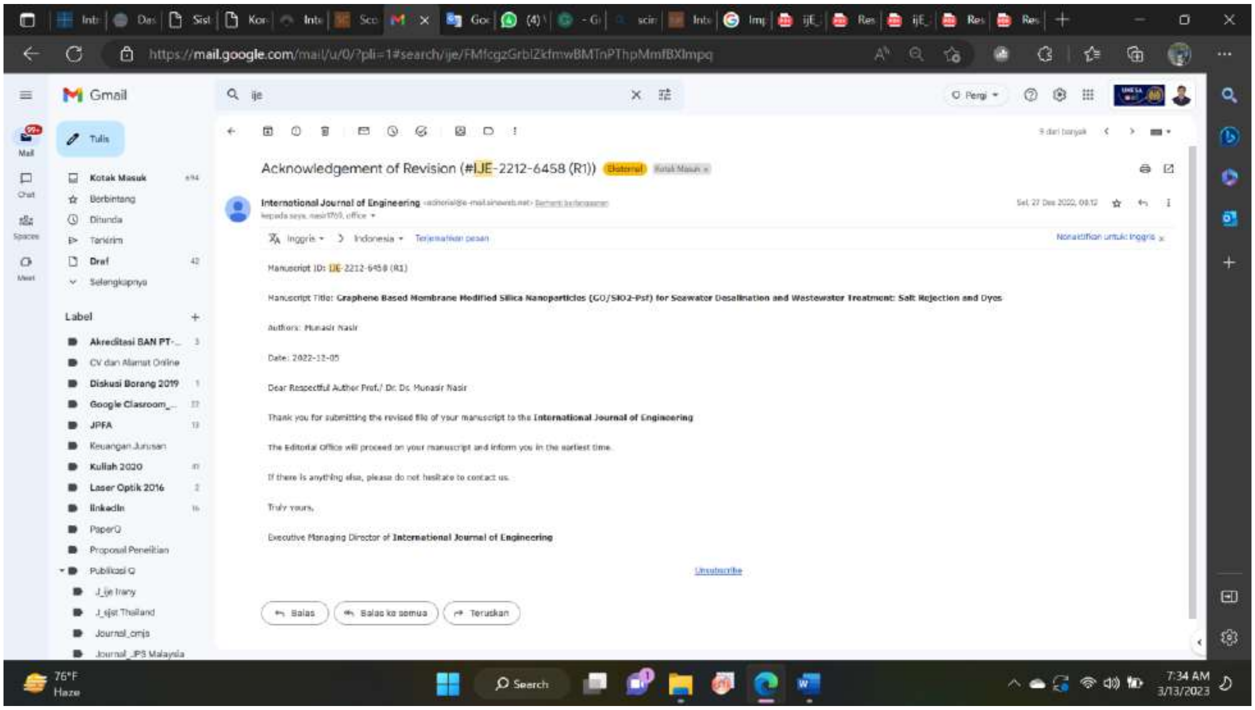

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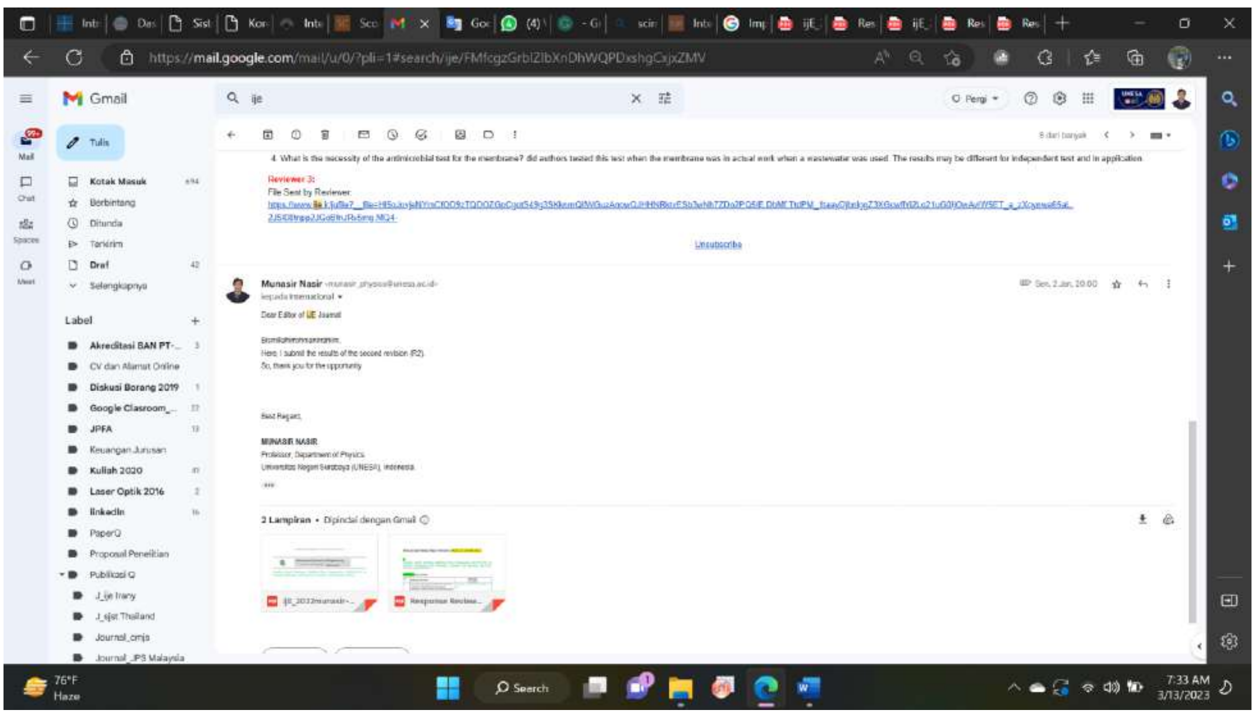

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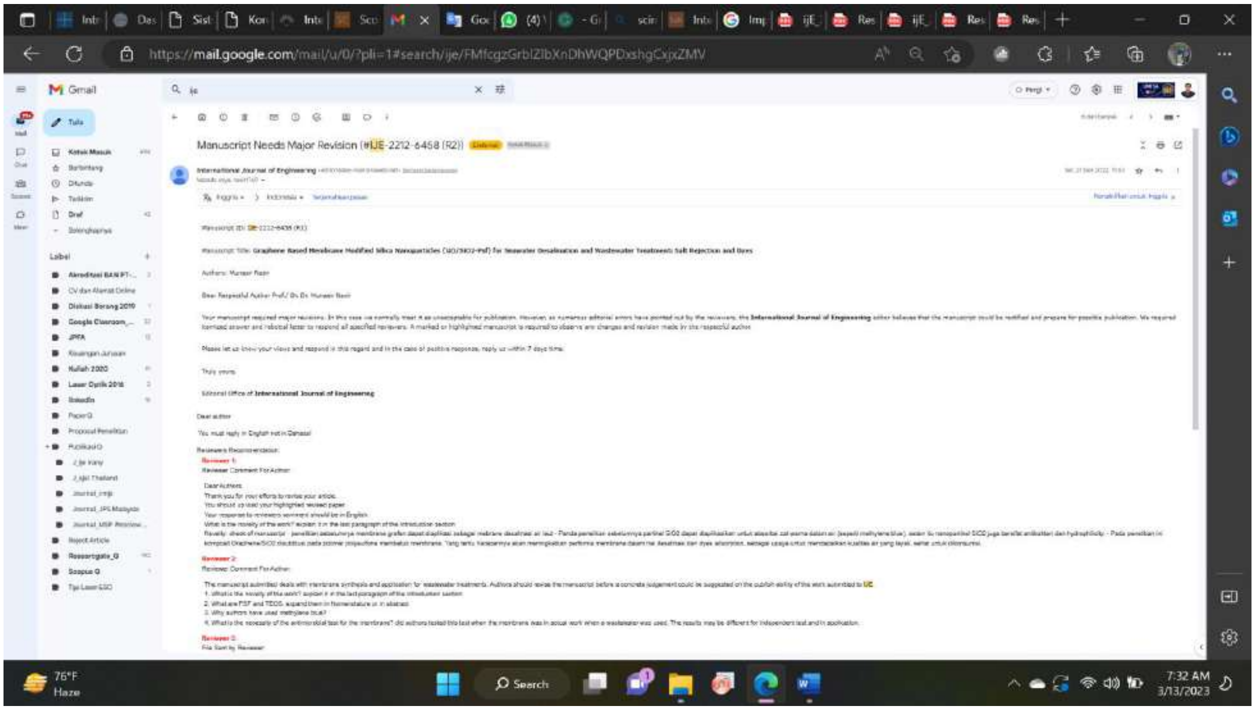
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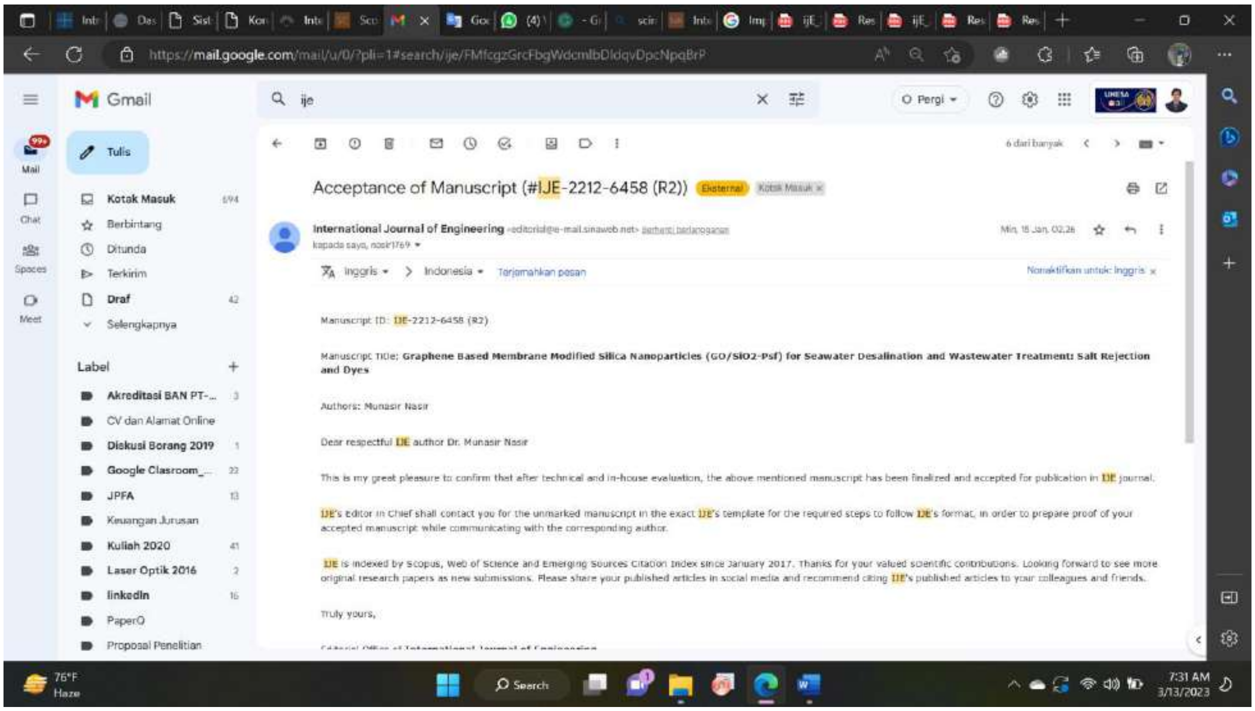
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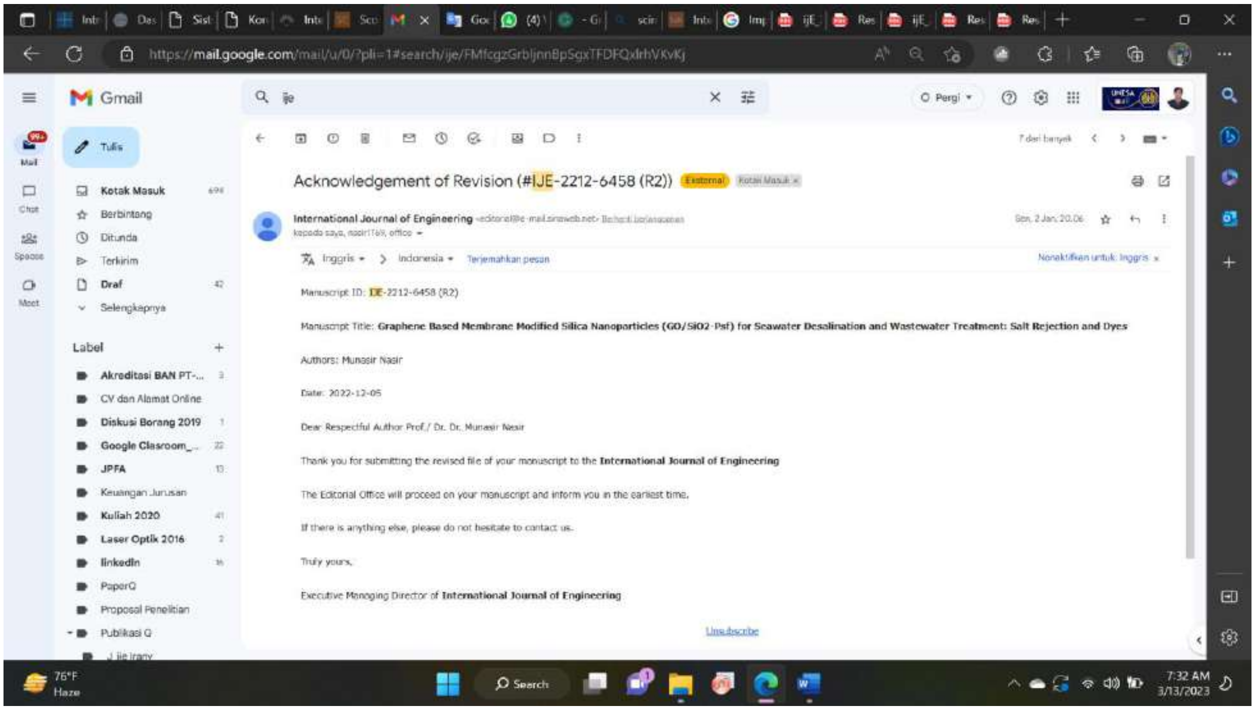

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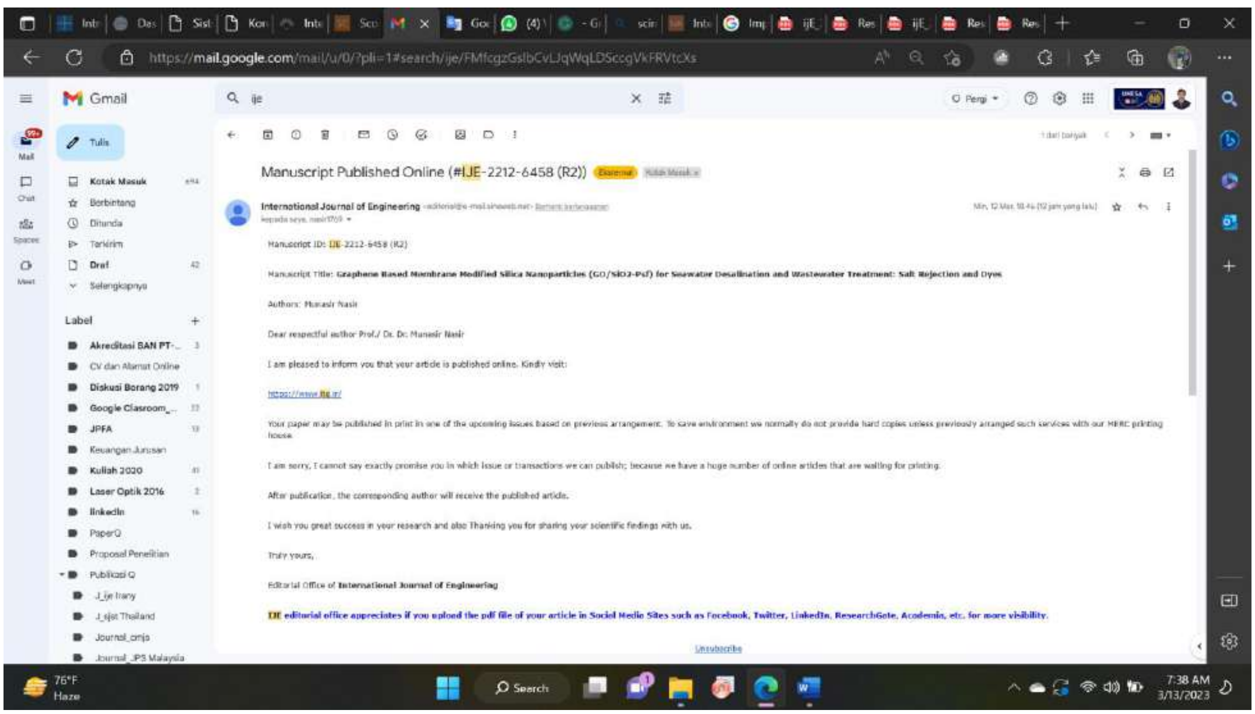

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Graphene Based Membrane Modified Silica Nanoparticles (GO/SiO_2-Psf) for Seawater Desalination and Wastewater Treatment: Salt Rejection and Dyes

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A B S T R A C T

The clean water crisis in Indonesia is increasing every year, and waste from the textile industry sector can also add to this problem. There are various water treatment processes to deal with the clean water crisis, one of which is the desalination process using graphene oxide. With the addition of hydrophilic nanoparticles, graphene oxide (GO) membranes can increase roughness and have good mechanical strength. SiO2 nanoparticles also have a high specific surface to absorb water or are hydrophilic. This study aims to determine the ability of the GO-SiO2/Psf membrane to reject salt (NaCl solution) and filtering of methylene blue solutions. Membrane prepared by variations of TEOS 0.6; 0.8; 1.0; and 1.2 ml for GO/SiO2 composite synthesis. The results showed that the GO-SiO2/PSF membrane could absorb methylene blue solution. The most optimum absorption value occurred at a TEOS concentration of 0.8 ml and had the most effective salt rejection value for NaCl solution equal to 67.22%.

***doi**: 10.5829/ije.2018.31.04a.01*

NOMENCLATURE			
C_f	The salt concentrations in feed	A	Area (m^2)
C_P	The salt concentrations in permeate	SR	Salt-Rejection (%)
J_w	The permeate water flux ($L\ m^{-2}\ h^{-1}$)	F	Flux flow
V	Volume (m^3)	A	The water permeability coefficient ($L\ m^{-2}\ h^{-1}\ bar^{-1}$)
TEOS	Tetraethyl orthosilicate, $Si(OH)_4$	Psf	Polysulfones , performance thermoplastics

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1. Introduction

Indonesia is the largest archipelagic country in the world with a sea area of 5,8 million km^2 and only 2,01 million km^2 of land area. By having an existing sea area, Indonesia has great potential to be able to utilize and treat sea water as an alternative to meet the needs of clean water. The need for clean water in Java from 1991 to 2020 was 79,41 billion m^3 and is predicted to increase by 7,24 billion m^3 from 2021 to 2050, while the availability of clean water in Java is in the range from 1991 to 2020 and 2021 to 2020. By 2050, it will decrease by 75,68 billion m^3 (1).

The scarcity of clean water is caused by several factors, including the amount of water demand that continues to increase and exceeds its supply capacity (2), as well as the result of irresponsible human activities (3,4). The scarcity of

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clean water also has an impact on the lower middle-class community because they have to provide funds to meet the needs of clean water. Global water scarcity is driven by water quantity and water quality issues, and measures expansion in clean water technologies (i.e. desalination and reuse of treated wastewater) "to reduce the number of people suffering from water scarcity" as urgently needed by the United Nations Sustainable Development Goal (SDGs 6.0) (5).

Meanwhile, the textile industry sector has increased every year. The textile industry can produce approximately 700 tons of dyes per year globally (6). Methylene blue is one of the dyes used in the textile industry as a basic ingredient in production. Methylene blue is a heterocyclic aromatic compound that is often used in the textile, silk, and wool industries (7). This can lead to the release of dye waste that endangers the surrounding environment and can damage aquatic organisms (8). Based on data from the World Resources Institute (WRI), Indonesia is ranked 51st with a high-risk level of clean water crisis (high 40–80% possibility). To deal with the scarcity of clean water, there are various kinds of water treatment processes, one of which is the desalination process.

Desalination is a water purification technique by separating the levels of substances from water (9,10) by reducing ions to the required level according to human needs (11). The desalination process can be carried out using graphene oxide. Graphene oxide offers an unusually high surface area, mechanical durability, atomic thickness, nano-sized pores and reactivity to polar and non-polar water pollutants. These characteristics provide high selectivity and water permeability, and thus provide excellent water purification efficiency. It also has the ability to adsorb and photocatalyzed water pollutants, so it has great potential for filtration materials, even for seawater desalination(9).

The addition of graphene oxide (GO) to the composite can affect the magnetization value so that it has super paramagnetic properties and can be used to absorb methylene blue (12,13). The GO membranes contain groups such as epoxide, carboxyl, and hydroxyl which can bind to water. Graphene oxide membranes also have good mechanical strength, so they are easy to fabricate and have the potential to be produced on an industrial scale (14-16). However, pure GO membranes have a finely stacked structure and have limited improvement in membrane performance. Based on the Cassie-Wenzel theory, there is an effective method to overcome the weakness of GO membranes, namely the addition of hydrophilic nanoparticles to increase surface roughness (17).

There are three graphene derivatives: graphite, GO, and rGO. Graphite is the primary material of carbon which is amorphous and stacked and rich in carbon and oxygen. GO is the result of graphite oxidation which increases the amount of oxygen, and some Van der Walls bonds have been released. So the thickness of the sample has been reduced. Reduced Graphene Oxide (rGO) is a graphene oxide in which the carbon atoms of graphene undergo oxidation and reduction. In the oxidation process, there are several oxygen and hydrogen atoms bonded to carbon atoms, the result of this oxidation process is called GO. While in the reduction process, some hydrogen and oxygen bonds are released from graphene oxide so that a structure almost similar to graphene is obtained.

From various previous studies, silica nanoparticles have shown great application potential in some fields, such as chemistry, biomedicine, biotechnology, agriculture, environmental improvement, and wastewater purification. With superior properties such as mesoporous structure, high surface area, adjustable pore size/diameter, morphology, biocompatibility, modifiability, anti-bacterial, as an excellent encapsulating agent for various bioactive molecules: proven safe for targeted drug delivery, and polymer hybridization ability (18).

In a study conducted by Jiawei Sun et al. using a GO-SiO_2 membrane as an oil-in-water separator, it was found that the incorporation of SiO_2 nanoparticles with GO can expand and increase water permeability with oil rejection (>99%) for various types of oil-in-water emulsions (17, 19–21). The GO/SiO_2 hybrid composite membrane has good hydrophilicity and thermal properties, able to reject high rhodamine B dye molecules (99%), high permeation and water resistance, so it is very good to be developed as a high-performance material for water treatment (22). SiO_2-GO/Psf hybrid membrane presents the best overall properties, including water permeation rate, protein rejection and antifouling ability (23,24). The SiO_2-GO nanohybrid has high hydrophilicity and good dispersibility properties derived from silica nanoparticles which are densely and uniformly coated on the GO surface and serve as a space layer of GO (22,23). The GO-SiO_2/Psf composite under UV irradiation during filtration greatly reduces the formation of fouling and produces a high flux recovery ratio, and is effective for filtration and remove organic pollutants (25).

SiO_2 nanoparticles have properties that can carry a large number of hydroxyl groups, and the concentration of hydroxyl groups is directly proportional to the specific surface area of the amorphous silica. And also have a high specific surface so that they can absorb water or are hydrophilic, and on the surface of SiO_2 there are abundant siloxane groups (Si-O-Si) that can bridge oxygen atoms (26,27). In this study, we will report the results of the synthesis of GO/SiO_2 nanocomposite and the fabrication of the GO/SiO_2-Psf membrane and its antibacterial and hydrophobic properties as well as its potential as water desalination, and filtration of methylene blue molecules in water.

2. Materials and Method

2.1. Materials

Some of the materials used include graphite powder produced from coconut shell extract, $NaNO_3$(Merck, for analysis), $KMnO_4$ (Merck, for analysis), H_2O, H_2O_2 (Merck, 30%), NH_4OH (Merck, 25%), distilled water, and TEOS which are all obtained from Edu Lab, H_2SO_4 (Merck, 95-97%) was obtained from Indofa Industry, HCl (Merck, 37%) was obtained from Mallinckrodt, NMP solvent was obtained from Sigma-Aldrich, PSF (Polysulfone) was obtained from Sigma-Aldrich (average Mw ~35,000), dan Methylene blue (M9140, Sigma-Aldrich).

2.2. Synthesis Method

2.2.1. Synthesis of Graphene (GO)

The hummer method was used, in which 5 grams of graphite powder, 2,5 grams of $NaNO_3$, and 120 ml of H_2SO_4 were placed in a 500 ml beaker with an ice bath and stirred for 30 minutes to form a black solution. Then 15 grams of $KMnO_4$ were added slowly and stirred for 30 minutes at a controlled temperature of 20°C to form a purple solution, and then stirred again for 3 hours at room temperature to form a brown solution. Then 150 ml of H_2O was added and the temperature was held at 95°C. The mixture was stirred for 3 hours at a controlled temperature of 95°C-100°C to form a dark yellow solution (28). Then 50 ml of H_2O_2 (30%) was added slowly, then washed with HCl (1M) and H_2O until the yellow color of the solution disappeared and the pH of the solution became neutral, then sonicated for 1 hour and filtered to produce a black gel. Furthermore, the black gel was dried at a temperature of 60°C for 6 hours and produced GO.

2.2.2. Synthesis of GO/SiO_2 Composite

It was carried out by the TEOS hydrolysis method, where 12,5 mg of GO was mixed with 150 ml of ethanol distilled water using a ratio of 1:5 and then sonicated for 30 minutes. Then, ammonia was added until the PH of the mixture was close to 9. Then 0.6 ml to 1.2 ml of TEOS concentration were added to the solution and stirred for 30 minutes. Then it was stirred at room temperature for 24 hours, centrifuged and washed with ethanol, then dried for 12 hours at 60°C producing a GO/SiO_2 composite (27).

2.2.3. Fabrication of GO/SiO_2-Psf Membrane

Using the phase inversion method, 1 gram of polysulfone was added to 5,469 ml of NMP and stirred for 3 hours. Then 0,0333 gram of GO/SiO_2 composite was added to obtain a 0.5wt% solution and sonicated for 30 minutes to shorten the dissolution process. No air bubbles were formed in it. Then the solution was formed on a glass plate and soaked in distilled water for 24 hours. Then it was dried for 24 hours at room temperature, which finally produced a GO/SiO_2-Psf membrane (29).

2.3. Characterizations

2.3.1. Characterization of GO/SiO_2 and GO/SiO_2-Psf Membrane

Phase and crystal structure analysis of nanoparticles (GO, SiO_2) and GO/SiO_2 nanocomposites used X-Ray Diffraction (PANalytical X'Pert Pro) plus Expert High Score Plus software; using monochromatic radiation CuKα (40kV/40 mA) taken at an angle of $5° \leq 2\theta \leq 90°$ at a rate of 0.02°/min at room temperature. Functional group analysis using infrared wave absorption test using FTIR (Shimadzu Brand, Type: IR-Prestige 21). Lattice vibration analysis in Graphene phase using Ramman spectroscopy (wavelength range 4000-200 cm^{-1}), and to observe the morphology of GO/SiO_2 nanocomposite and GO/SiO_2-Psf membrane using the Scanning electron microscope/SEM test (FEI Brand, Type: Inspect-S50). UV-Visible test to analyze the adsorption of methylene blue dye in water. And the analysis of the hydrophilicity of the membrane surface with the CAA 2320 Contact Angle instrument.

2.3.4. Filtration of NaCl Solution

GO/SiO_2-Psf was placed in a funnel Buchner flask and a hose was connected to a vacuum pump (Rocker 300) (Fig. 1). The vacuum pump was turned on, then the membrane was dripped with 10 ml of NaCl solution (as sea-water synthetic) at a pressure of 650 mmHg for all variations of the GO/SiO_2-Psf membrane. The membrane that has been passed through the NaCl solution is then weighed.

2.3.5. Filtration of Methylene Blue in Aqueous

The 20 ppm methylene blue solution was passed through a 12,25 cm^2 $GO\text{-}SiO_2/PSF$ membrane placed in a Buchner flask funnel and connected to a vacuum pump (Rocker 300) using a tube (Fig.1). The vacuum pump was turned on, then the membrane was dripped with 10 ml of methylene blue solution at a pressure of 650 mmHg for all variations of the $GO\text{-}SiO_2/PSF$ membrane. The results of the filtered methylene blue solution will be subjected to a UV-Visible test to determine the absorbance (30). The results of the filtered methylene blue solution will be tested by UV-Vis (Shimadzu 1800) with a wavelength of 200-600 nm, to determine the absorption.

Methylene blue (C16H18N3SCl) is one of the most commonly used dyes for dyeing wool, cotton and silk. Since sewage or water containing dyes can cause serious environmental problems and the availability of healthy water for consumption, it is necessary to treat these wastes before being discharged into the environment (31).

2.3.6. Salt-Rejection Test

A salt-rejection test was carried out to determine the efficiency of the membrane in filtering NaCl solution. Salt-rejection (SR) can be obtained by the following formula (11):

$$Salt - rejection = \frac{Cf - Cp}{Cf} x100 \qquad (1)$$

Where, *Cf* is the mean salt concentration in feed stream, and *Cp* is the salt concentration in the permeate.

2.3.5. Flow-Flux Test

The membrane flux test was carried out to measure the optimization parameters of the membrane. The flow flux can be obtained by the formula (11):

$$J = \frac{V}{At} \qquad (2)$$

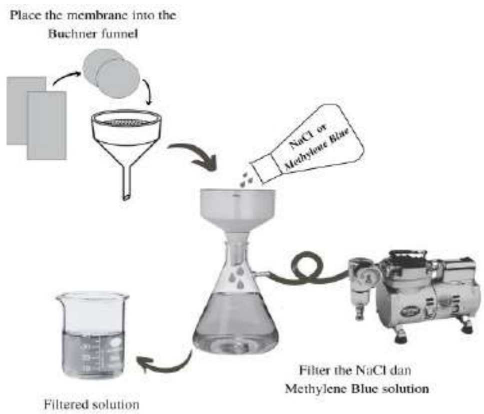

Figure 1. Filtration test with GO/SiO_2-Psf

3. Result And Discussion

3.1. Pattern Diffraction of GO and Composite

Diffraction analysis of Graphene (GO) material and GO/SiO_2 composite is presented in Fig. 2. The peak at 2θ= 10.21° is associated with the (002) GO plane in the GO/SiO_2 composite, but this peak does not appear. It is associated with weak diffraction, and the presence of graphite oxide heaps, in which SiO_2 is coated with a GO sheet, causes GO diffraction to appear (23). The similarity of the diffraction peaks for GO and the GO/SiO_2 composite is indicated at positions 2θ =21.8° and 26.2°. And the presence of a widened peak at 2θ = 23.31° indicates that silica is amorphous (30).

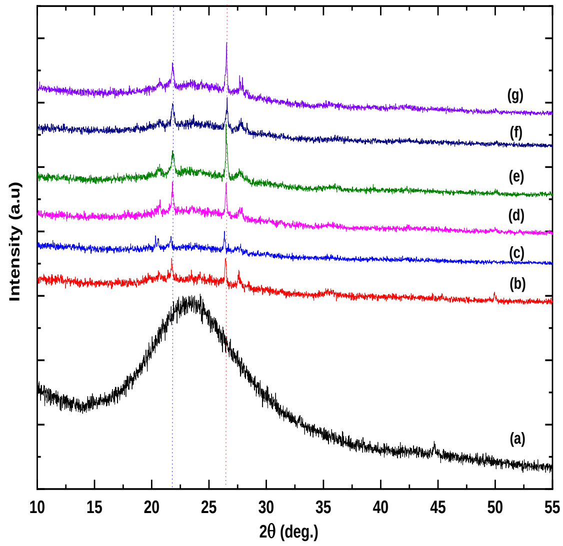

Figure 2. Pattern Diffraction of SiO_2(a), Graphite(b), GO (c) and GO/SiO_2-Psf Composite (d-g)

3.2. Functional Group of GO and Composite

The functional group analysis of the characteristics for GO materials and GO/SiO_2 composites is presented in Fig. 3. The results of the analysis showed the presence of silanol groups and Si-O silicates and C-H carbon groups. These functional groups are a representation of the composite material (13,33). The GO/SiO_2 composite exhibits a new peak at 1100 cm^{-1}, characteristic of the Si-O-Si asymmetric vibration. The presence of this absorption proves that silica is on the GO surface (23). In addition, the peak at 1600-1735 cm^{-1} is associated with the C=O vibration of the carboxylate group. The decreasing C=O absorption peak in the composite was due to the interaction of C=O with GO to become Si-O-C indicating a bonding interaction between GO and Silica.

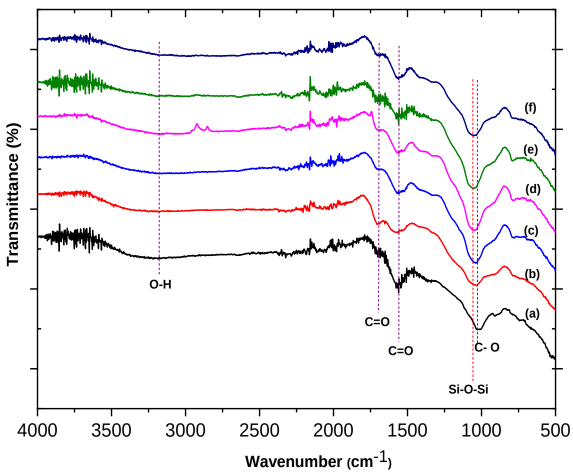

Figure 3. Functional group of GO and GO/SiO_2-Psf: (a) Grafite, (b) GO, dan (c-f) composite.

3.3. Morphology of GO, GO/SiO_2 Composite and GO/SiO_2-Psf Membrane

The morphology of GO/SiO_2 composite material and GO/SiO_2-Psf membrane with NPM solvent during the membrane preparation process is presented in Fig. 4. The shape of the particles varies, it appears that the particles have several (small) pores, and between the stacks of particle arrangement there are quite large voids. Particle sizes also vary, small and large, this affects the formation of voids which will later play a role in forming the density of the membrane material.

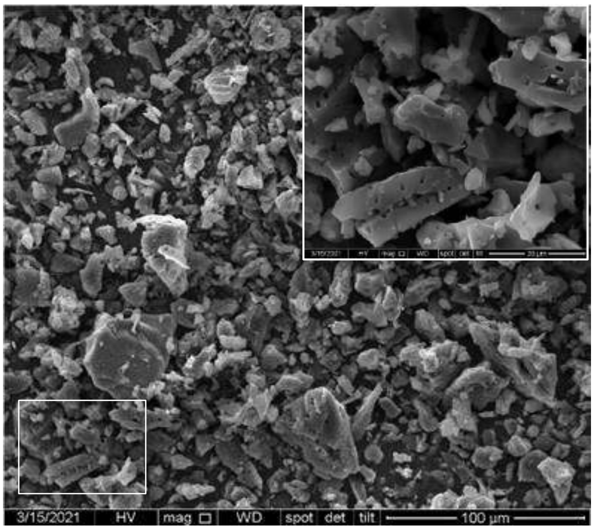

Figure 4. Morphology of GO/SiO_2 composite

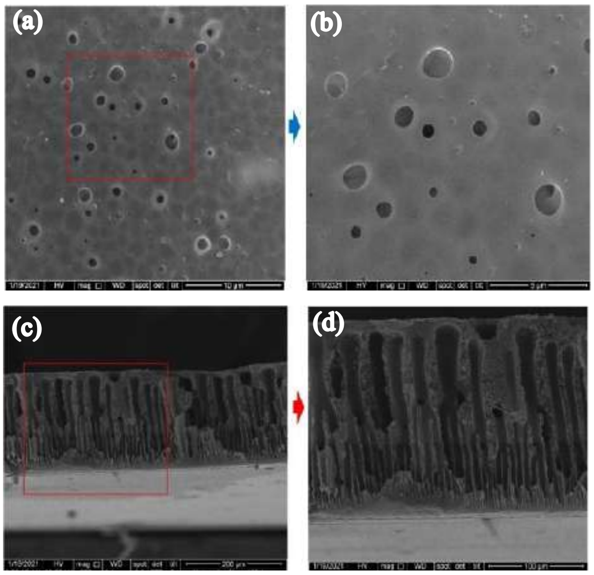

Figure 5. Morphology of GO/SiO_2-Psf membrane: (a-b) *surface pores*, (c-d) *cross-sectional view*.

The morphology of the GO/SiO_2-Psf membrane material prepared by the phase inversion method is presented in Fig. 5. It can be seen on the surface of the membrane and its cross section. On the surface of the membrane, pores of varying sizes were seen, and these were identified as water inlet channels through the membrane through the cavities formed on the inside of the membrane. The size and shape of the cavity is influenced by the type of membrane polymer (polysulfon) and the composite material embedded in the membrane.

3.4. Hydrophobicity of GO/SiO_2-Psf Membrane

The contact angle of GO/SiO_2-Psf polysulfone membrane with NMP solvent is presented as follows, for the weight percent of GO/SiO_2 composite (for SiO_2, *Wt%* ≈ 0.5%) with the percentage of composite material varied: 0.6; 0.8; 1.0; and 1.2%. Fig.5 and Table 1 show that the angle is smaller than 90° (hydrophobic): each membrane sample shows an average contact angle of: 75.41°; 70.16°; 76.78°; and 74.25°. On the other hand, for the SiO2-Psf membrane contact angle (without GO) the contact angle is 71.13°, approximately the same as the GO/SiO_2-Psf membrane. This indicates that the membrane is hydrophilic or slightly water-loving (contact angle (θ) (0°≤θ≤90°) (33,34).

3.5. Anti-Bacterial of GO/SiO_2 Composite

The disc diffusion test was carried out to determine the inhibitory power of the compound on bacterial growth. Bacterial suspension (∅D600 nm 0.1) was rubbed on the surface of Muller Hinton Agar (MHA) media on a Petri dish using a sterile cotton swab. A paper disk containing 20 l of the test compound was placed on the surface of the MHA. Incubation was carried out for 48 hours at 30ºC. The clear zone formed around the disc was expressed as the inhibitory power of the compound against bacterial growth. Anti-bacterial test in this study, using 2 types of bacteria, namely Escherichia coli and Staphylococcus aureus.

The results of the antimicrobial test of GO/SiO_2 against Escherichia coli and Staphylococcus aureus with the disc diffusion method (Table 2) showed that the inhibitory power of E. Coli was stronger than S. Aureus; The increase in GO-SiO2 (ppm) concentration further strengthens the antibacterial activity of the E. Coli strain, compared to S. aureus which tends to remain stable. Thus, the membrane material will be confirmed to have resistance to bacterial growth (E. Coli and S. Aureus) (34). Bacterial-mediated infections can cause various acute or chronic diseases and antibiotic resistance in pathogenic bacteria has become a serious health problem. Graphene-based materials have been very well studied due to their outstanding bactericidal activity on various bacteria. The use of GO material in membrane preparation will provide biosafety advantages (35). Likewise, the presence of SiO_2 nanoparticles, also has excellent properties as an anti-bacterial material (36,37).

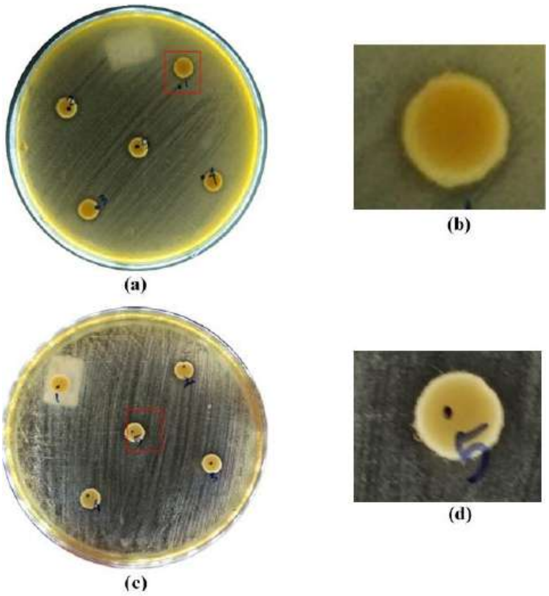

Figure 6. Photograph of antimicrobial of GO/SiO_2: (a-b) E. Coli and (c-d) S. Aureus

3.6. Salt-Rejection of NaCl Solution

Based on Table 1, the results of the salt-rejection calculation, the Cp value is obtained from the total solid NaCl dissolved in freshwater with a salt density of 2,16 g/ml, and the Cf value is obtained from the initial concentration in the NaCl solution, which is 27,79 g/ml. The GO/SiO_2-Psf membrane with a TEOS (0,8 ml) showed the most effective results for seawater desalination because the GO/SiO_2-Psf membrane was able to filter out NaCl compounds in solution by 67,22%. The high salt-rejection value is caused by the distribution of silica grains on the membrane being very tight so that the pores are getting smaller and able to filter NaCl compounds efficiently (15). The GO/SiO_2-Psf membrane with TEOS concentrations of 1,0 ml and 1,2 ml decreased because the ratio of matrix and filler was not balance.

TABLE 1. Contact Angle of GO/SiO_2-Psf(0.5%) Membrane with DMAC and NMP Solvents

PSF-0,5% GO/SiO_2	*Contact Angle (degrees) with DMAC solvent*			*Contact Angle (degrees) with NMP solvent*		
	Right	*Left*	*Average*	*Right*	*Left*	*Average*
0,60 ml TEOS	76,13	81,02	***78,58***	74,39	76,44	***75,41***
0,80 ml TEOS	61,78	60,36	***61,07***	69,71	70,61	***70,16***
1,00 ml TEOS	82,71	82,53	***82,62***	72,65	80,90	***76,78***
1,20 ml TEOS	70,24	70,86	***70,55***	74,79	73,72	***74,25***
0,5% SiO_2-PSF	69,91	72,44	***71,18***	70,43	71,84	***71,13***

TABLE 2. Antimicrobial of GO-SiO_2 compounds against E. coli and S. Aureus

Bacterial	***test repeats***	***Inhibition zone diameter (cm) at various concentrations of GO-SiO_2 compounds***				
		100 ppm	200 ppm	300 ppm	400 ppm	500 ppm
Escherichia coli	1	0,83	0,94	1,01	1,08	0,97
	2	0,79	0,83	0,91	0,95	1,04
	3	0,93	0,94	0,94	0,98	1,01
Staphylococcus aureus	1	0,55	0,55	0,55	0,55	0,55
	2	0,55	0,55	0,55	0,55	0,55
	3	0,55	0,55	0,55	0,55	0,55

3.6. Filtration of Methylene Blue in Aqueous

By using 20 ppm methylene blue was then carried out a filtration test. The methylene blue solution was filtered using a GO/SiO_2-Psf membrane with various concentrations of TEOS 0,6; 0,8; 1,0; and 1,2 ml. The results of the methylene blue solution filtration test (Fig.2) show that the methylene blue solution, which has been filtered using a GO/SiO_2-Psf membrane, appears to change in color before and after filtering.

The UV-Vis test was carried out using a wavelength of 200 to 600 nm, and the absorption value of each sample was obtained (Fig.3). The methylene blue solution without passing through the GO-SiO_2/Psf membrane obtained an absorbance value of 1,206 at a wavelength of 291 nm. Figure 7 shows the results of the UV-Vis test showing the relationship between wavelength and absorbance. The UV-Vis absorption spectrum of GO/SiO_2 nanocomposite has a strong absorption in the range of 200-650 nm. In the composite for TEOS (0.6 ml), GO showed a characteristic peak at 325 nm, for TEOS (0.8 ml) it decreased starting at the peak of 280 nm. For TEOS (1.0 and 1.2 ml) indicates a stable position and continues to increase. The maximum absorption peak corresponds to the π-π* transition bond in the C=C aromatic structure and the $\pi \rightarrow \pi$* transition in the carbonyl group (C=O). The characteristic peak shifts to 270 nm for GO due to the presence of reduced graphene (22,40).

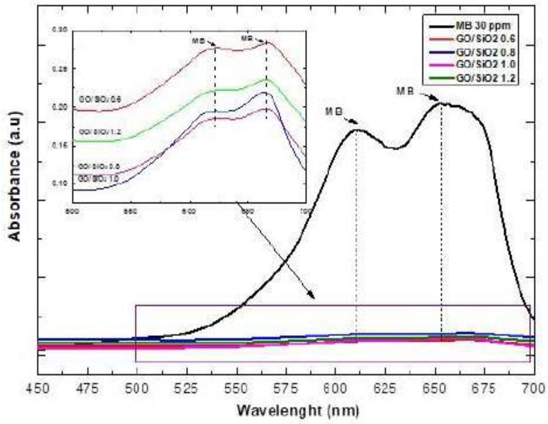

Figure 7. Dye absorption in water (Methylene Blue) by GO/SiO_2 nanocomposite.

The Methylene blue, which was passed through the GO/SiO_2-Psf membrane in one time filtering (Fig. 8a) decreased the absorption value, where the most optimum absorbance occurred in the membrane with a TEOS concentration of 0,8 ml, which was 0,396 at a wavelength of 291 nm. This is to the literature conducted by Nurul et al, 2019 where the absorbance of methylene blue can be carried out using silica (**17).** This also supports the statement of Cahyo et al, 2018 that the higher the silica content used, the larger the surface area, so the more effective it is to absorb methylene blue (20). However, on membranes with TEOS concentrations of 1,0 ml and 1,2 ml, there was an increase in the absorbance value due to the unbalanced and uneven matrix and filler content during the manufacture of the membrane, which caused the membrane to be less than optimal in the filtering process.

Based on Fig. 8b, the methylene blue solution that has been passed through the GO/SiO_2-Psf membrane 5 times has been filtered. There is a significant decrease in the absorbance value compared to the 1 filter (Table 3). The more the filtering process uses the GO/SiO_2-Psf membrane, the lower the methylene blue content in the solution. By the statement of Neldawati, 2013 the content of methylene blue in the solution that has been passed through the membrane decreases when the filtration test is completed (21).

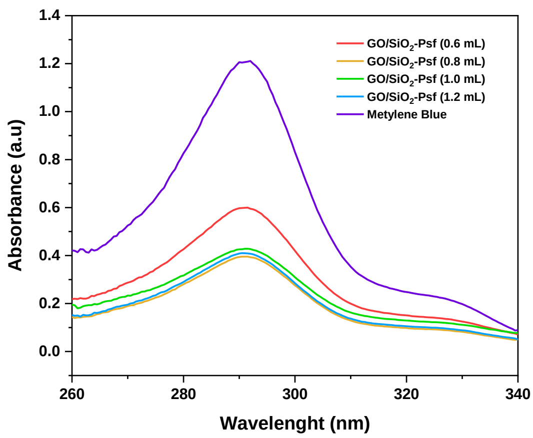

Figure 8. UV-Vis test of Methylene Blue solution 1 times filtering

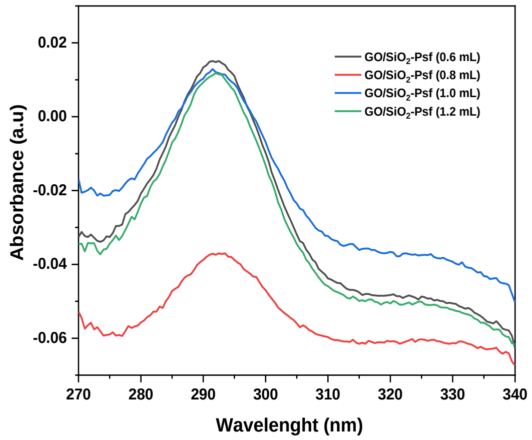

Figure 9. UV-Vis test of Methylene Blue solution 5 times filtering

Visually (Fig. 10) there is a significant color change, and it is relatively clearer when the color change is by the results of the UV-Vis test that has been carried out. The GO/SiO2-Psf membrane with TEOS 0,8 ml at 5 times of filtration obtained a negative absorbance value (Table 3), which indicates that the sample does not contain methylene blue analyte or that the methylene blue content contained in the solution is below the detection limit of the UV spectrometer method. So, it can be said that the addition of SiO_2 to the membrane can maximize the membrane as an absorbent (22), and the GO-SiO2/PSF membrane with 0,8 ml TEOS is more effective in filtering methylene blue solutions.

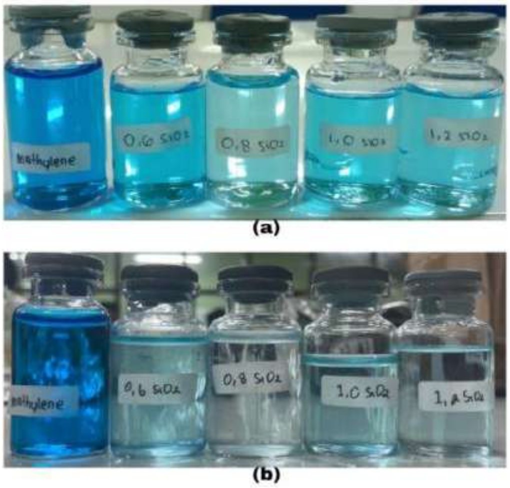

Figure 10. Metylene Blue solution filtration test results (a) 1 filter, (b) 5 filters.

Table 3. Comparison of absorbance values of Methylene Blue solution one-times filtering and fives-times filtering

Wt% *Membrane*	*Absorbance (a.u)*	
	1 times filtering	*5 times filtering*
GO/SiO_2 *(0,6)*	*0,599*	*0,015*
GO/SiO_2 *(0,8)*	*0,396*	*-0,037*
GO/SiO_2 *(1,0)*	*0,428*	*0,012*
GO/SiO_2 *(1,2)*	*0,409*	*0,011*

3.7. Flow Flux of NaCl Solution

Based on the experimental results, the data obtained are as in table 3. The magnitude of the flow flux value is affected for different GO-SiO2/PSF membrane samples, the greater the composition of SiO^2 in the flow-flux membrane the greater. The value of the flow flux for NaCl solution and methylene blue water can be seen in Table 3. A low flux value indicates a low membrane permeability, so it can be said that the membrane is more optimal in filtering the solution, the resulting water quality is cleaner (from impurities), especially salt molecules. and methylene-blue molecules (natural dyes).

The filtering time of the solution greatly affects the value of the resulting water flux, where the longer the filtering time, the GO/SiO_2-Psf membrane has a tight pore size. The GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml had the lowest flux value, namely 47,46 $L.m^{-2}.h^{-1}$ in NaCl solution and 51,55 $L.m^{-2}.h^{-1}$ in methylene blue ocean. The decrease in flux in the membrane can be caused by the closure of several pores in the membrane. The membrane flux will decrease as the filtering time increases (17-19, 41). The GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml has a tight pore size so that it is more efficient for filtering NaCl and methylene blue solutions.

Table 4. The results of the calculation of flow flux

Wt% *Membrane*	*Time (Hour)*		*Flux ($L.m^{-2}.h^{-1}$)*	
	NaCl (sea-water synthetic)	*Metylene Blue*	*NaCl (sea-water synthetic)*	*Metylene Blue*
GO/SiO_2 *(0,6)*	0,13	0,12	79,37	81,30
GO/SiO_2 *(0,8)*	0,17	0,16	47,46	51,55
GO/SiO_2 *(1,0)*	0,15	0,14	53,71	59,88
GO/SiO_2 *(1,2)*	0,16	0,15	48,88	54,05

3.8. Flow Flux of NaCl Solution and Methylene-Blue in Water

Fig. 11(a) is the result of one filter where the methylene blue solution is still concentrated. Fig. 11(b) is the result of 5 times filtering, where the methylene blue solution is relatively clearer. Visually, it can be seen that the GO/SiO_2-Psf membrane can reduce or absorb methylene blue gradually. Based on the results of the filtration test on the methylene blue solution 5 times filtering, the GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml looked the brightest, it was by the flux test that had been carried out.

The filtering time of the solution greatly affects the value of the resulting water flux, where the longer the filtering time, the GO/SiO_2-Psf membrane has a tight pore size. The GO/SiO_2-Psf (0.8 ml) membrane had the lowest flux value, namely 47,46 $L.m^{-2}.h^{-1}$ in NaCl solution and 51,55 L.m-2.h-1 in methylene blue ocean. The decrease in flux in the membrane can be caused by the closure of several pores in the membrane. The membrane flux will decrease as the filtering time increases (42). The GO/SiO_2-Psf (0.8 ml) membrane has a tight pore size so that it is more efficient for filtering NaCl and methylene blue solutions.

In addition, the graphene structure that forms thin sheets of net (the order of nanometers) is very effective for filtering water molecules. GO modified with SiO2 in the membrane system is believed to be more effective in the filtration and absorption of pollutants in water (Fig. 11). Graphene oxide (GO) has unique characteristics that make it an excellent material for water purification applications. Chemically stable in water, provides high water permeability through its 2D nanochannels, and has excellent antifouling and antibacterial properties (43).

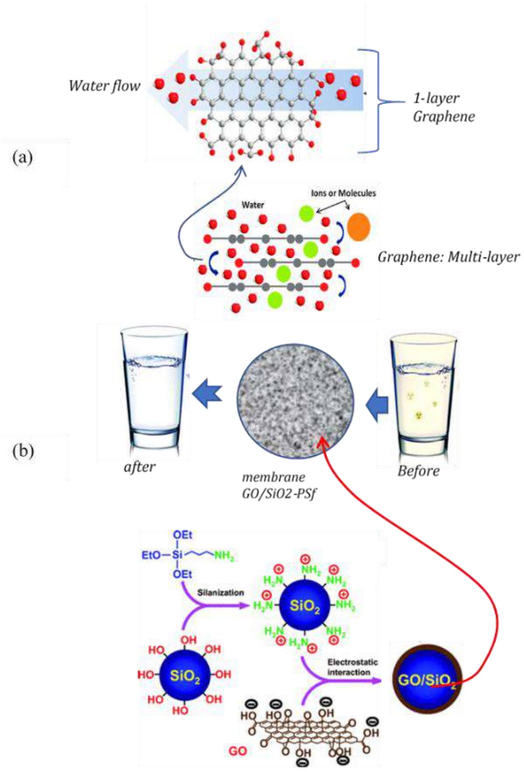

Figure 11. Filtration model of GO/SiO_2-Psf membrane: (a) water-flow in layer graphene, (b) fresh-water after filtrations.

Graphene has unique physicochemical properties, extraordinarily high surface area, mechanical resistance, atomic thickness, nano-sized pores, and polar reactivity of polar and non-polar water, thus providing high selectivity and water permeability and thus providing excellent water purification efficiency. Graphene material has great potential as a membrane for water desalination, GO for good adsorption, and photocatalysis of water pollutants. On the other hand, SiO2 nanoparticles have good adsorption properties due to their high surface area and porosity, so they can be promoted as membrane fillers (40, 43-46).

Synthetic polymers, such as polysulfone, have been successfully used in water and wastewater treatment due to their unique chemical and physical characteristics, such as high chemical resistance, mechanical properties, and thermal stability. These polymeric materials tend to be hydrophobic by default and are prone to organic fouling, which requires doping with hydrophilic monomers/materials to induce hydrophilic properties to overcome fouling problems. Low-energy plasma and irradiation are used, and hydrophilic organic and inorganic materials can be mixed in the polymer by irradiation (8,43).

4. Conclusions

The GO/SiO_2 composite material can resist the growth of bacteria (E. Coli and S. Aureus). GO/SiO_2-Psf membrane can be applied well to absorb color molecules in water (methylene blue) and has the most effective salt rejection value for salt solution (desalination process). Membranes with SiO_2 composition (~0.8 ml) showed natural dyes absorbing and desalination performance (salt-rejection up to 67.22%). The high salt rejection value and low absorbance value are caused by the distribution of silica grains on the membrane so that the pores are getting smaller and can filter NaCl and methylene blue molecules very well.

Acknowledgments

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Manuscript Needs Major Revision (#IJE-2212-6458 (R1))

R1- **Graphene/SiO2-Psf Membrane for Water-Filtration: Seawater (NaCl) and Dyes Methylene Blue (MB)-** Manuscript ID: IJE-2212-6458

#1Editorial EJE:

No	Editor IJE Comments	Responses (for author)
1	Please cite IJE's recent published papers which are related to your research area	*(1)* IJE TRANSACTIONS A: Basics Vol. 33, No. 1, (January 2020) 18-27: *Characterization of Fe_3O_4/rGO Composites from Natural Sources: Application* ***for Dyes Color Degradation in Aqueous Solution*** *(2)* IJE TRANSACTIONS A: Basics Vol. 35, No. 05, (July 2022) 1300-1306: ***Synthesis of Silica Nanoparticles from Silica Sand*** *via Vibration Assisted Alkaline Solution Method* *(3)* IJE TRANSACTIONS A: Basics Vol. 32, No. 7, (July 2019) 982-990: Corrosion Polarization Behavior of Al-**SiO_2** Composites in 1M and Related Microstructural Analysis *(4)* IJE TRANSACTIONS A: Basics Vol. 30, No. 10, (October 2017) 1425-1433: Preparation and Characterization of an Antifouling Polyethersulfone **Nanofiltration Membrane** Blended with Graphene Oxide/Ag Nanoparticles *(5)* IJE TRANSACTIONS C: Aspects Vol. 29, No. 9, (September 2016) 1191-1197: **Adsorption of Methylene Blue from Aqueous Solutions** by Silk Cocoon
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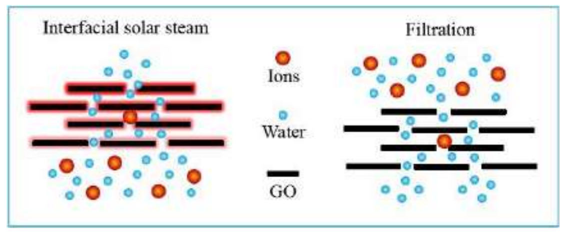
No	Referee's Comments	Responses (for author)
1	The manuscript submitted deals with membrane synthesis and application for wastewater treatments. Authors should revise the manuscript before a concrete judgement could be suggested on the publish-ability of the work submitted to IJE.	Okay, thank you. we will do soon
2	What is the novelty of the work? explain it in the last paragraph of the Introduction section	**Novelty: *check of manuscript*** - penelitian sebelumnya membrane grafen dapat diaplikasi sebagai mebrane desalinasi air laut - Panda penelitian sebelumnya partikel SiO2 dapat diaplikasikan untuk absorbsi zat warna dalam air (seperti methylene blue), selain itu nanopartikel SiO2 juga bersifat antibakteri dan hydrophilicity. - Pada penelitian ini komposit Graphene/SiO2 disubtitusi pada polimer polysulfone membetuk membrane. Yang tentu harapannya akan meningkatkan performa membrane dalam hal desalinasi dan dyes adsorption, sebagai upaya untuk mendapatkan kualitas air yang layak, sehat untuk dikonsumsi.
3	What are PSF and TEOS. expand them in Nomenclature or in abstract	- PSF = **Polysulfones** are a family of high performance thermoplastics. These polymers are known for their toughness and stability at high temperatures - TEOS= Tetraethyl orthosilicate (***$Si(OH)_4$***), as a precursor to silicon dioxide, silica nanoparticles. ***Check in nomenclature***
4	Why authors have used methylene blue?	**Methylene blue (C16H18N3SCl) is one of the most commonly used dye** stuff for dying wool, cotton and silk. Since dye-containing effluents can cause serious environmental problems, they need to be treated before being discharged into the surroundings. The treatment processes for removal of dye from water and

		wastewater can be grouped into three categories: biological (i.e. using microorganisms like bacteria, yeasts, algae and fungi), chemical (i.e. coagulation, precipitation-flocculation, oxidation, electrochemical processes), and ***physical (i.e. membrane-filtration, adsorption) [N. S. Tabrizi, 2016]*** For the dye color in the water adsorption test, several types of dyes (synthetic or natural) can be used; however, methylene blue is used to facilitate ***identification by UV-Visible spectroscopy***.
5	What is the necessity of the antimicrobial test for the membrane? did authors tested this test when the membrane was in actual work when a wastewater was used. The results may be different for independent test and in application.	The reason is that this membrane will be used as a medium for the filtration of contaminated water or desalination so that that water can be consumed healthily. So, one must also be ensured that the membrane material (Graphen/SiO2) has good anti-bacterial properties. Graphene/SiO2 material testing is carried out before use (in actual work). It is intended for initial performance tests before technical application and after wastewater filtration tests. Agree that the results may vary for independent testing and after application.

#3Reviewer2: minor revision

No	Referee's Comments	Responses (for author)
1	From the title, it is not easy to understand the content of the study very well. So it is suggested to change it.	Previous title: ***Graphene/SiO2-Psf Membrane for Water-Filtration: Sea-Water (NaCl) and Dyes Methylene Blue (MB)*** New title: ***Graphene Based Membrane Modified Silica Nanoparticles (GO/SiO2-Psf) for Seawater and Wastewater Treatment: Salt Rejection and Dyes***
2	In the abstract; concentrate on your findings and achievements. If the authors are discussing their study should make it specific and clear and more detailed.	**New abstract**: The clean water crisis in Indonesia is increasing every year, and waste from the textile industry sector can also add to this problem. There are various water treatment processes to deal with the clean water crisis, one of which is the desalination process using graphene oxide. With the addition of hydrophilic nanoparticles, graphene oxide (GO) membranes can increase roughness and have good mechanical strength. SiO2 nanoparticles also have a high specific surface to absorb water or are hydrophilic. This study aims to determine the ability of the GO-SiO2/Psf membrane to reject salt (NaCl solution) and filtering of methylene blue solutions. Membrane prepared by variations of TEOS 0.6; 0.8; 1.0; and 1.2 ml for GO/SiO2 composite synthesis. The results showed that the GO-SiO2/PSF membrane could absorb methylene blue solution. The most optimum absorption value occurred at a TEOS concentration of 0.8 ml and had the most effective salt rejection value for NaCl solution equal to 67.22%.
3	The used methods, investigated parameters, characterization methods and techniques are all common ones, and one can hardly see any novelty in them. Can you mention the novelty of this study and advantages over other methods?	Check in manuscript, **Material and Method:** Synthesis of Graphene, Silica Nanoparticles (SiO2), or GO/SiO2 composites, as well as Graphene (GO) membranes are commonly carried out by previous researchers. The novelty is that GO/SiO2 becomes the basis of nanocomposite as a membrane filler (polysulfone, which is thermoplastic), resulting in a graphene-based membrane with

		modification of silica nanoparticles. Nanoparticles fill between GO sheets. The basic idea is that filtered water passes between the GO sheets, and SiO2 particles which are inert, porous and anti-bacterial are positioned between the GO sheets. Thus, in addition to getting pure water (H2O) with very little impurities carried, it can also ensure that it is clean from micro-organisms such as bacteria and viruses. And the final product is water that is ready, very suitable and healthy for drinking water. **The Illustrations [Xiuqiang, 2019]:**  *Notes:* 1. *The d-spacing of nanochannels (GO) is about 0.76 nm, which is larger than water molecules (0.275 nm).* 2. *The ions (Li^+ (0.764 nm), Na^+ (0.716 nm), K^+ (0.662 nm), Mg^{2+} (0.856 nm), Ca^{2+} (0.824 nm), F^- (0. 704 nm), Cl^- (0.664 nm), Br^- (0.660 nm)) can be separated from seawater through size exclusion and electrostatic interactions, thus achieving desalination GO film filtration*
4	Page 2, column 2, line 3 correct the spelling of Graphene.	Page 2, column 2, line 3 correct the spelling of Graphene. ***Check on manuscript***
5	Section 2-3-4 what does " Cf dan Cp berturut-turut adalah konsentrasi" mean?	Section 2-3-4: Cf is the mean salt concentration in feed stream, and Cp is the salt concentration in the permeate.
6	Section 2-3-4 define the parameters of the equations.	**Section 2-3-4:** Cf is the mean salt concentration in feed stream, and Cp is the salt concentration in the permeate.
7	Provide references for peaks appeared in XRD patterns.	Reference: peak position (hkl) in XRD data, Figure2: ***Check on manuscript***

8	The figures are not numbered correctly in the text.	Thanks, I corrected
9	Page 8, column 2, the paragraph "there are 3 derivatives …." should be moved to the introduction part.	Page 8, column 2, the paragraph "there are 3 derivatives …." should be moved to the introduction part→**Check in manuscript (in introduction)**
10	For how long can the membrane be used optimally?	In this initial test, the filtration test with water NaCl and MB for a single-layer membrane arrangement, yet to be in a spinning membrane such as technical applications. Single sheets have shown good functional trends. So, with the technical application, it will be much more effective. Thus, the membrane can function appropriately for a standard time.

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Graphene Based Membrane Modified Silica Nanoparticles (GO/SiO_2-Psf) for Seawater Desalination and Wastewater Treatment: Salt Rejection and Dyes

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A B S T R A C T

The clean water crisis in Indonesia is increasing every year, and waste from the textile industry sector can also add to this problem. There are various water treatment processes to deal with the clean water crisis, one of which is the desalination process using graphene oxide. With the addition of hydrophilic nanoparticles, graphene oxide (GO) membranes can increase roughness and have good mechanical strength. SiO2 nanoparticles also have a high specific surface to absorb water or are hydrophilic. This study aims to determine the ability of the GO-SiO2/Psf membrane to reject salt (NaCl solution) and filtering of methylene blue solutions. Membrane prepared by variations of TEOS 0.6; 0.8; 1.0; and 1.2 ml for GO/SiO2 composite synthesis. The results showed that the GO-SiO2/PSF membrane could absorb methylene blue solution. The most optimum absorption value occurred at a TEOS concentration of 0.8 ml and had the most effective salt rejection value for NaCl solution equal to 67.22%.

doi: *10.5829/ije.2018.31.04a.01*

NOMENCLATURE			
C_f	The salt concentrations in feed	*A*	Area (m^2)
C_P	The salt concentrations in permeate	*SR*	Salt-Rejection (%)
J_w	The permeate water flux ($L\ m^{-2}\ h^{-1}$)	*F*	Flux flow
V	Volume (m^3)	A	The water permeability coefficient ($L\ m^{-2}\ h^{-1}\ bar^{-1}$)
TEOS	Tetraethyl orthosilicate, $Si(OH)_4$	Psf	Polysulfones , performance thermoplastics

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1. Introduction

Indonesia is the largest archipelagic country in the world with a sea area of 5,8 million km^2 and only 2,01 million km^2 of land area. By having an existing sea area, Indonesia has great potential to be able to utilize and treat sea water as an alternative to meet the needs of clean water. The need for clean water in Java from 1991 to 2020 was 79,41 billion m^3 and is predicted to increase by 7,24 billion m^3 from 2021 to 2050, while the availability of clean water in Java is in the range from 1991 to 2020 and 2021 to 2020. By 2050, it will decrease by 75,68 billion m^3 (1).

The scarcity of clean water is caused by several factors, including the amount of water

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demand that continues to increase and exceeds its supply capacity (2), as well as the result of irresponsible human activities (3,4). The scarcity of clean water also has an impact on the lower middle-class community because they have to provide funds to meet the needs of clean water. Global water scarcity is driven by water quantity and water quality issues, and measures expansion in clean water technologies (i.e. desalination and reuse of treated wastewater) "to reduce the number of people suffering from water scarcity" as urgently needed by the United Nations Sustainable Development Goal (SDGs 6.0) (5).

Meanwhile, the textile industry sector has increased every year. The textile industry can produce approximately 700 tons of dyes per year globally (6). Methylene blue is one of the dyes used in the textile industry as a basic ingredient in production. Methylene blue is a heterocyclic aromatic compound that is often used in the textile, silk, and wool industries (7). This can lead to the release of dye waste that endangers the surrounding environment and can damage aquatic organisms (8). Based on data from the World Resources Institute (WRI), Indonesia is ranked 51st with a high-risk level of clean water crisis (high 40–80% possibility). To deal with the scarcity of clean water, there are various kinds of water treatment processes, one of which is the desalination process.

Desalination is a water purification technique by separating the levels of substances from water (9,10) by reducing ions to the required level according to human needs (11). The desalination process can be carried out using graphene oxide. Graphene oxide offers an unusually high surface area, mechanical durability, atomic thickness, nano-sized pores and reactivity to polar and non-polar water pollutants. These characteristics provide high selectivity and water permeability, and thus provide excellent water purification efficiency. It also has the ability to adsorb and photocatalyzed water pollutants, so it has great potential for filtration materials, even for seawater desalination(9).

The addition of graphene oxide (GO) to the composite can affect the magnetization value so that it has super paramagnetic properties and can be used to absorb methylene blue (12,13). The GO membranes contain groups such as epoxide, carboxyl, and hydroxyl which can bind to water. Graphene oxide membranes also have good mechanical strength, so they are easy to fabricate and have the potential to be produced on an industrial scale (14-16). However, pure GO membranes have a finely stacked structure and have limited improvement in membrane performance. Based on the Cassie-Wenzel theory, there is an effective method to overcome the weakness of GO membranes, namely the addition of hydrophilic nanoparticles to increase surface roughness (17).

SiO_2 nanoparticles have properties that can carry a large number of hydroxyl groups, and the concentration of hydroxyl groups is directly proportional to the specific surface area of the amorphous silica. And also have a high specific surface so that they can absorb water or are hydrophilic, and on the surface of SiO2 there are abundant siloxane groups (Si-O-Si) that can bridge oxygen atoms (18, 19). From this characteristic, there are many advantages, such as the broad application of biomedical and biotechnological applications, agricultural applications, industrial applications, environmental applications, and water purification. In water purification, SN can reduce biological oxygen demand, perform antimicrobial strength as a filter for water-oil mixture, and filter methylene red, commonly used in the textile and paper industry, as waste disposal from a textile factory.

There are three graphene derivatives: graphite, GO, and rGO. Graphite is the primary material of carbon which is amorphous and stacked and rich in carbon and oxygen. GO is the result of graphite oxidation which increases the amount of oxygen, and some Van der Walls bonds have been released. So the thickness of the sample has been reduced. Reduced Graphene Oxide (rGO) is a graphene oxide in which the carbon atoms of graphene undergo oxidation and reduction. In the oxidation process, there are several oxygen and hydrogen atoms bonded to carbon atoms, the result of this oxidation process is called GO. While in the reduction process, some hydrogen and oxygen bonds are released from graphene oxide so that a structure almost similar to graphene is obtained.

From various previous studies, silica nanoparticles have shown great application potential in some fields, such as chemistry, biomedicine, biotechnology, agriculture, environmental improvement, and wastewater purification. With superior properties such as mesoporous structure, high surface area, adjustable pore size/diameter, morphology, biocompatibility, modifiability, anti-bacterial, as an excellent encapsulating agent for various bioactive molecules: proven safe for targeted drug delivery, and polymer hybridization ability (20).

In a study conducted by Jiawei Sun et al. using a GO-SiO_2 membrane as an oil-in-water separator, it was found that the incorporation of SiO_2 nanoparticles with GO can expand and increase water permeability with oil rejection (>99%) for various types of oil-in-water emulsions (17, 21–23). The GO/SiO_2 hybrid composite membrane has good hydrophilicity and thermal properties, able to reject high rhodamine B dye molecules (99%), high permeation and water resistance, so it is very good to be developed as a high-performance material for water treatment (24). SiO_2-GO/Psf hybrid membrane presents the best overall properties, including water permeation rate, protein rejection and antifouling ability (25,26). The SiO_2-GO nanohybrid has high hydrophilicity and good dispersibility properties derived from silica nanoparticles which are densely and uniformly coated on the GO surface and serve as a space layer of GO (24,25). The GO-SiO_2/Psf composite under UV irradiation during filtration greatly reduces the formation of fouling and produces a high flux recovery ratio, and is effective for filtration and remove organic pollutants (27).

Previous studies on graphene membranes have shown good performance as seawater desalination membranes (9). Furthermore, silica nanoparticles can be applied as adsorption of dyes in water (such as methylene blue); In addition, SiO_2 nanoparticles also have good antibacterial and hydrophilic properties. In this study, substituted graphene silica nanoparticles and polysulfone polymers were fabricated into membranes using the phase inversion method. This study is expected to improve the performance of membranes in desalination and adsorption of dyes to obtain decent water quality, healthy for consumption. GO/SiO_2-Psf membrane performance in salt rejection (for desalination) and dye filtration in water (MB) will be presented in this report.

2. Materials and Method

2.1. Materials

Some of the materials used include graphite powder produced from coconut shell extract, $NaNO_3$(Merck, for analysis), $KMnO_4$ (Merck, for analysis), H_2O, H_2O_2 (Merck, 30%), NH_4OH (Merck, 25%), distilled water, and TEOS which are all obtained from Edu Lab, H_2SO_4 (Merck, 95-97%) was obtained from Indofa Industry, HCl (Merck, 37%) was obtained from Mallinckrodt, NMP solvent was obtained from Sigma-Aldrich, PSF (Polysulfone) was obtained from Sigma-Aldrich (average Mw ~35,000), dan Methylene blue (M9140, Sigma-Aldrich).

2.2. Synthesis Method

2.2.1. Synthesis of Graphene (GO)

The hummer method was used, in which 5 grams of graphite powder, 2,5 grams of $NaNO_3$, and 120 ml of H_2SO_4 were placed in a 500 ml beaker with an ice bath and stirred for 30 minutes to form a black solution. Then 15 grams of $KMnO_4$ were added slowly and stirred for 30 minutes at a controlled temperature of 20°C to form a purple solution, and then stirred again for 3 hours at room temperature to form a brown solution. Then 150 ml of H_2O was added and the temperature was held at 95°C. The mixture was stirred for 3 hours at a controlled temperature of 95°C-100°C to form a dark yellow solution (28). Then 50 ml of H_2O_2 (30%) was added slowly, then washed with HCl (1M) and H_2O until the yellow color of the solution disappeared and the pH of the solution became neutral, then sonicated for 1 hour and filtered to produce a black gel. Furthermore, the black gel was dried at a temperature of 60°C for 6 hours and produced GO.

2.2.2. Synthesis of GO/SiO_2 Composite

It was carried out by the TEOS hydrolysis method, where 12,5 mg of GO was mixed with 150 ml of ethanol distilled water using a ratio of 1:5 and then sonicated for 30 minutes. Then, ammonia was added until the PH of the mixture was close to 9. Then 0.6 ml to 1.2 ml of TEOS concentration were added to the solution and stirred for 30 minutes. Then it was stirred at room temperature for 24 hours, centrifuged and washed with ethanol, then dried for 12 hours at 60°C producing a GO/SiO_2 composite (27).

2.2.3. Fabrication of GO/SiO_2-Psf Membrane

Using the phase inversion method, 1 gram of polysulfone was added to 5,469 ml of NMP and stirred for 3 hours. Then 0,0333 gram of GO/SiO_2 composite was added to obtain a 0.5wt% solution and sonicated for 30 minutes to shorten the dissolution process. No air bubbles were formed in it. Then the solution was formed on a glass plate and soaked in distilled water for 24 hours. Then it was dried for 24 hours at room temperature, which finally produced a GO/SiO_2-Psf membrane (29).

2.3. Characterizations

2.3.1. Characterization of GO/SiO_2 and GO/SiO_2-Psf Membrane

Phase and crystal structure analysis of nanoparticles (GO, SiO_2) and GO/SiO_2 nanocomposites used X-Ray Diffraction (PANalytical X'Pert Pro) plus Expert High Score Plus software; using monochromatic radiation CuKα (40kV/40 mA) taken at an angle of 5°≤2θ_≤90° at a rate of 0.02°/min at room temperature. Functional group analysis using infrared wave absorption test using FTIR (Shimadzu Brand, Type: IR-Prestige 21). Lattice vibration analysis in Graphene phase using Ramman spectroscopy (wavelength range 4000-200 cm^{-1}), and to observe the morphology of GO/SiO_2 nanocomposite and GO/SiO_2-Psf membrane using the Scanning electron microscope/SEM test (FEI Brand, Type: Inspect-S50). UV-Visible test to analyze the adsorption of methylene blue dye in water. And the analysis of the hydrophilicity of the membrane surface with the CAA 2320 Contact Angle instrument.

2.3.4. Filtration of NaCl Solution

GO/SiO_2-Psf was placed in a funnel Buchner flask and a hose was connected to a vacuum pump (Rocker 300) (Fig. 1). The vacuum pump was turned on, then the membrane was dripped with 10 ml of NaCl solution (as sea-water synthetic) at a pressure of 650 mmHg for all variations of the GO/SiO_2-Psf membrane. The membrane that has been passed through the NaCl solution is then weighed.

2.3.5. Filtration of Methylene Blue in Aqueous

The 20 ppm methylene blue solution was passed through a 12,25 cm^2 GO-SiO_2/PSF membrane placed in a Buchner flask funnel and connected to a vacuum pump (Rocker 300) using a tube (Fig.1). The vacuum pump was turned on, then the membrane was dripped with 10 ml of methylene blue solution at a pressure of 650 mmHg for all variations of the GO-SiO_2/PSF membrane. The results of the filtered methylene blue solution will be subjected to a UV-Visible test to determine the absorbance (30). The results of the filtered methylene blue solution will be tested by UV-Vis (Shimadzu 1800) with a wavelength of 200-600 nm, to determine the absorption.

Methylene blue (C16H18N3SCl) is one of the most commonly used dyes for dyeing wool, cotton and silk. Since sewage or water containing dyes can cause serious environmental problems and the availability of healthy water for consumption, it is necessary to treat these wastes before being discharged into the environment (31).

2.3.6. Salt-Rejection Test

A salt-rejection test was carried out to determine the efficiency of the membrane in filtering NaCl solution. Salt-rejection (SR) can be obtained by the following formula (11):

$$Salt - rejection = \frac{Cf - Cp}{Cf} x100 \qquad (1)$$

Where, Cf is the mean salt concentration in feed stream, and Cp is the salt concentration in the permeate.

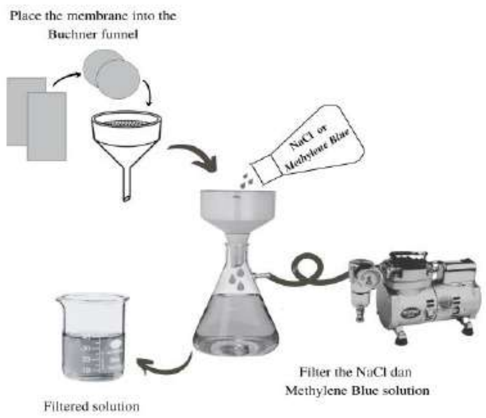

Figure 1. Filtration test with GO/SiO_2-Psf

2.3.5. Flow-Flux Test

The membrane flux test was carried out to measure the optimization parameters of the membrane. The *filtrate flow flux* (J) can be obtained by the formula (11):

$$J = \frac{V}{At} \qquad (2)$$

Where A is membrane area (m^2), V is the volume of filtrate generated (liter), and t is process time (hours).

3. Result And Discussion

3.1. Pattern Diffraction of GO and Composite

Diffraction analysis of Graphene (GO) material and GO/SiO_2 composite is presented in Fig. 2. The peak at 2θ= 10.21° is associated with the (002) GO plane in the GO/SiO_2 composite, but this peak does not appear. It is associated with weak diffraction, and the presence of graphite oxide heaps, in which SiO_2 is coated with a GO sheet, causes GO diffraction to appear (23). The similarity of the diffraction peaks for GO and the GO/SiO_2 composite is indicated at positions 2θ =21.8° and 26.2°. And the presence of a widened peak at 2θ = 23.31° indicates that silica is amorphous (30).

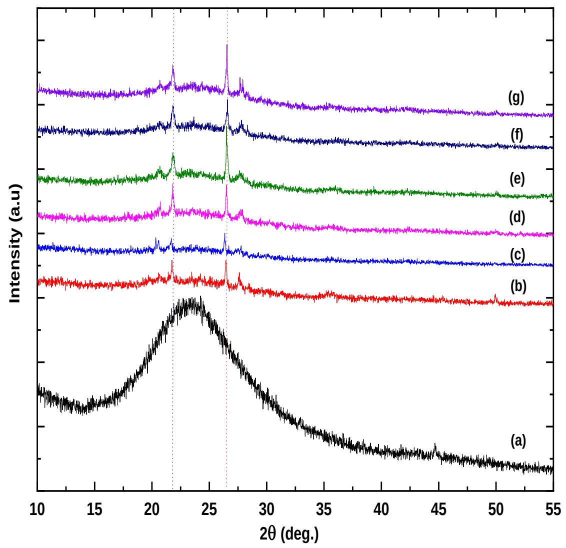

Figure 2. Pattern Diffraction of SiO_2(a), Graphite(b), GO (c) and GO/SiO_2-Psf Composite (d-g)

3.2. Functional Group of GO and Composite

The functional group analysis of the characteristics for GO materials and GO/SiO_2 composites is presented in Fig. 3. The results of the analysis showed the presence of silanol groups and Si-O silicates and C-H carbon groups. These functional groups are a representation of the composite material (13,33). The GO/SiO_2 composite exhibits a new peak at 1100 cm^{-1}, characteristic of the Si-O-Si asymmetric vibration. The presence of this absorption proves that silica is on the GO surface (23). In addition, the peak at 1600-1735 cm^{-1} is associated with the C=O vibration of the carboxylate group. The decreasing C=O absorption peak in the composite was due to the interaction of C=O with GO to become Si-O-C indicating a bonding interaction between GO and Silica.

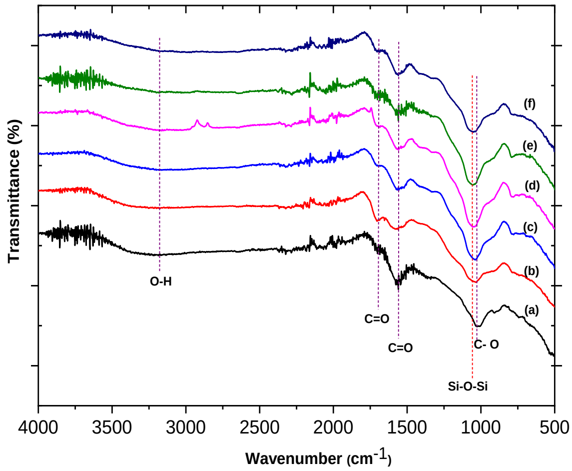

Figure 3. Functional group of GO and GO/SiO_2-Psf: (a) Grafite, (b) GO, dan (c-f) composite.

3.3. Morphology of GO, GO/SiO_2 Composite and GO/SiO_2-Psf Membrane

The morphology of GO/SiO_2 composite material and GO/SiO_2-Psf membrane with NPM solvent during the membrane preparation process is presented in Fig. 4. The shape of the particles varies, it appears that the particles have several (small) pores, and between the stacks of particle arrangement there are quite large voids. Particle sizes also vary, small and large, this affects the formation of voids which will later play a role in forming the density of the membrane material.

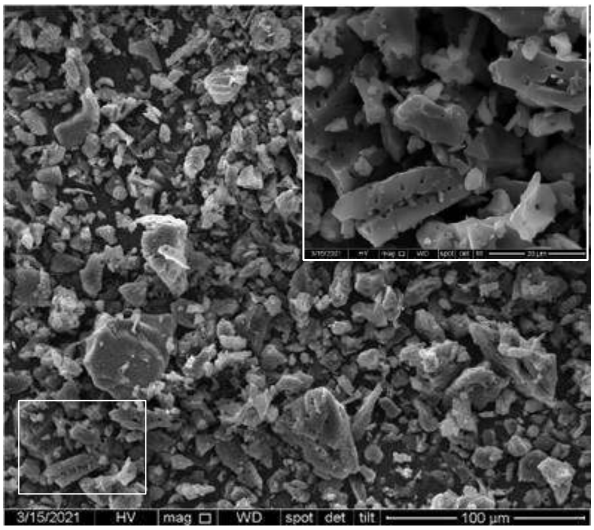

Figure 4. Morphology of GO/SiO_2 composite

The morphology of the GO/SiO_2-Psf membrane material prepared by the phase inversion method is presented in Fig. 5. It can be seen on the surface of the membrane and its cross section. On the surface of the membrane, pores of varying sizes were seen, and these were identified as water inlet channels through the membrane through the cavities formed on the inside of the membrane. The size and shape of the cavity is influenced by the type of membrane polymer (polysulfone) and the composite material embedded in the membrane. Figure 5b shows a cross-sectional view of the modified composite membrane with the topical asymmetrical morphology of the membrane fibers. The middle layer represents the predominant morphology with finger-like structures. This finger-like structure is characteristic of an asymmetrical membrane where the cross-section of the membrane consists of a finger structure with a porous underlayer (33). The formation of a porous surface on GO-SiO_2-psf is caused by an increase in the hydrophilic nature of the solution, which will accelerate the rate of solvent exchange (34). The addition of GO into the membrane causes a larger cavity. In theory, the more macro-voids appear, the greater the membrane's permeability (33).

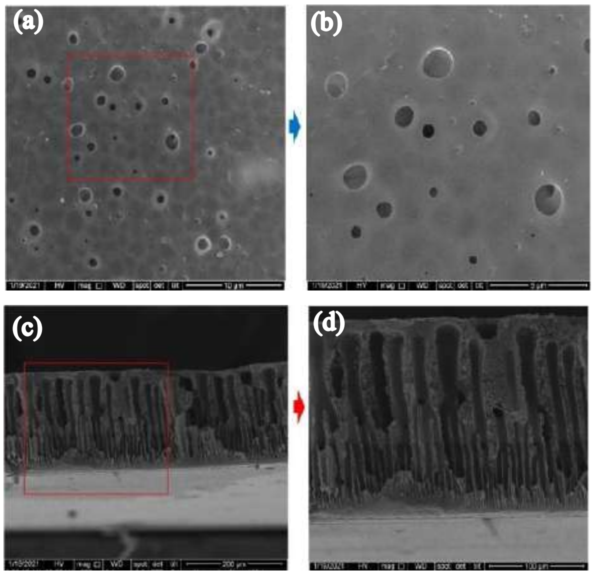

Figure 5. Morphology of GO/SiO_2-Psf membrane: (a-b) *surface pores*, (c-d) *cross-sectional view*.

3.4. Hydrophobicity of GO/SiO_2-Psf Membrane

The hydrophilic nature of the membrane has an essential role in filtration performance. In principle, the hydrophilicity of the membrane can be determined by the water contact angle.

The contact angle of GO/SiO_2-Psf polysulfone membrane with NMP solvent is presented as follows, for the weight percent of GO/SiO_2 composite (for SiO_2, $Wt\% \approx 0.5\%$) with the percentage of composite material varied: 0.6; 0.8; 1.0; and 1.2%. Fig.5 and Table 1 show that the angle is smaller than 90° (hydrophobic): each membrane sample shows an average contact angle of: 75.41°; 70.16°; 76.78°; and 74.25°. The lower the contact angle, the higher the membrane hydrophilicity (35; 36). The greater contact angle value is caused by the surface tension between the membrane and the water. On the other hand, for the SiO2-Psf membrane contact angle (without GO) the contact angle is 71.13°, approximately the same as the GO/SiO_2-Psf membrane. This indicates that the membrane is hydrophilic or slightly water-loving (contact angle (θ) (0°≤θ≤90°) (37,38). Furthermore, a small contact angle generally results in better hydrophilicity, increased water flux, and resistance to impurities (Guo et al., 2018). Thus, the experimental results obtained in this study are in accordance with the literature (40).

3.5. Anti-Bacterial of GO/SiO_2 Composite

The disc diffusion test was carried out to determine the inhibitory power of the compound on bacterial growth. Bacterial suspension (∅D600 nm 0.1) was rubbed on the surface of Muller Hinton Agar (MHA) media on a Petri dish using a sterile cotton swab. A paper disk containing 20 l of the test compound was placed on the surface of the MHA. Incubation was carried out for 48 hours at 30ºC. The clear zone formed around the disc was expressed as the inhibitory power of the compound against bacterial growth. Anti-bacterial test in this study, using 2 types of bacteria, namely Escherichia coli and Staphylococcus aureus.

The results of the antimicrobial test of GO/SiO_2 against Escherichia coli and Staphylococcus aureus with the disc diffusion method (Table 2) showed that the inhibitory power of E. Coli was stronger than S. Aureus; The increase in GO-SiO2 (ppm) concentration further strengthens the antibacterial activity of the E. Coli strain, compared to S. aureus which tends to remain stable. Thus, the membrane material will be confirmed to have resistance to bacterial growth (E. Coli and S. Aureus) (38). Bacterial-mediated infections can cause various acute or chronic diseases and antibiotic resistance in pathogenic bacteria has become a serious health problem. Graphene-based materials have been very well studied due to their outstanding bactericidal activity on various bacteria. The use of GO material in membrane preparation will provide biosafety advantages (41). Likewise, the presence of SiO_2 nanoparticles, also has excellent properties as an anti-bacterial material (42,43).

3.6. Salt-Rejection of NaCl Solution

Based on Table 4, the results of the salt-rejection calculation, the Cp value is obtained from the total solid NaCl dissolved in freshwater with a salt density of 2,16 g/ml, and the Cf value is obtained from the initial concentration in the NaCl solution, which is 27,79 g/ml. The GO/SiO_2-Psf membrane with a TEOS (0,8 ml) showed the most effective results for seawater desalination because the GO/SiO_2-Psf membrane was able to filter out NaCl compounds in solution by 67,22%. The high salt-rejection value is caused by the distribution of silica grains on the membrane being very tight so that the pores are getting smaller and able to filter NaCl compounds efficiently (15). The research of Zeng et al. (2019) showed that an increase in membrane thickness due to the pore density of SiO_2, can help reduce conductive losses in bulk water and increase salt rejection ability, but energy efficiency is limited by maximum liquid flux (44). The GO/SiO_2-Psf membrane with TEOS concentrations of 1,0 ml and 1,2 ml decreased because the ratio of matrix and filler was not balance.

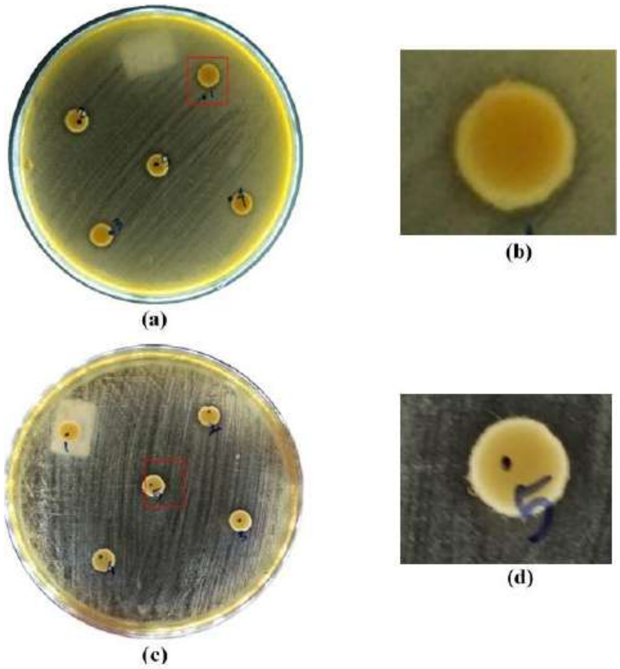

Figure 6. Photograph of antimicrobial of GO/SiO_2: (a-b) E. Coli and (c-d) S. Aureus

TABLE 1. Contact Angle of GO/SiO_2-Psf(0.5%) Membrane with DMAC and NMP Solvents

PSF-0,5% GO/SiO₂	*Contact Angle (degrees) with DMAC solvent*			*Contact Angle (degrees) with NMP solvent*		
	Right	*Left*	*Average*	*Right*	*Left*	*Average*
0,60 ml TEOS	76,13	81,02	***78,58***	74,39	76,44	***75,41***
0,80 ml TEOS	61,78	60,36	***61,07***	69,71	70,61	***70,16***
1,00 ml TEOS	82,71	82,53	***82,62***	72,65	80,90	***76,78***
1,20 ml TEOS	70,24	70,86	***70,55***	74,79	73,72	***74,25***
0,5% SiO_2-PSF	69,91	72,44	***71,18***	70,43	71,84	***71,13***

TABLE 2. Antimicrobial of GO-SiO_2 compounds against E. coli and S. Aureus

Bacterial	***test repeats***	***Inhibition zone diameter (cm) at various concentrations of GO-SiO₂ compounds***				
		100 ppm	200 ppm	300 ppm	400 ppm	500 ppm
Escherichia coli	1	0,83	0,94	1,01	1,08	0,97
	2	0,79	0,83	0,91	0,95	1,04
	3	0,93	0,94	0,94	0,98	1,01
Staphylococcus aureus	1	0,55	0,55	0,55	0,55	0,55
	2	0,55	0,55	0,55	0,55	0,55
	3	0,55	0,55	0,55	0,55	0,55

3.6. Filtration of Methylene Blue in Aqueous

By using 20 ppm methylene blue was then carried out a filtration test. The methylene blue solution was filtered using a GO/SiO_2-Psf membrane with various concentrations of TEOS 0,6; 0,8; 1,0; and 1,2 ml. The results of the methylene blue solution filtration test (Fig.2) show that the methylene blue solution, which has been filtered using a GO/SiO_2-Psf membrane, appears to change in color before and after filtering.

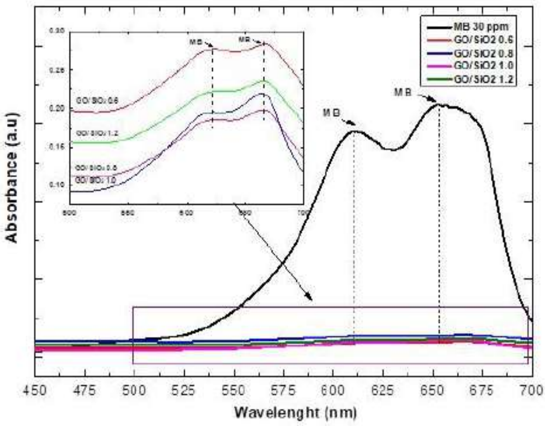

Figure 7. Dye absorption in water (Methylene Blue) by GO/SiO_2 nanocomposite.

The UV-Vis test was carried out using a wavelength of 200 to 600 nm, and the absorption value of each sample was obtained (Fig.3). The methylene blue solution without passing through the GO-SiO_2/Psf membrane obtained an absorbance value of 1,206 at a wavelength of 291 nm. Figure 7 shows the results of the UV-Vis test showing the relationship between wavelength and absorbance. The UV-Vis absorption spectrum of GO/SiO_2 nanocomposite has a strong absorption in the range of 200-650 nm. In the composite for TEOS (0.6 ml), GO showed a characteristic peak at 325 nm, for TEOS (0.8 ml) it decreased starting at the peak of 280 nm. For TEOS (1.0 and 1.2 ml) indicates a stable position and continues to increase. The maximum absorption peak corresponds to the π-π* transition bond in the C=C aromatic structure and the π→π* transition in the carbonyl group (C=O). The characteristic peak shifts to 270 nm for GO due to the presence of reduced graphene (22,47).

The Methylene blue, which was passed through the GO/SiO_2-Psf membrane in one time filtering (Fig. 8a) decreased the absorption value, where the most optimum absorbance occurred in the membrane with a TEOS concentration of 0,8 ml, which was 0,396 at a wavelength of 291 nm. This is to the literature conducted by Nurul et al, 2019 where the absorbance of methylene blue can be carried out using silica (**17).** This also supports the statement of Cahyo et al, 2018 that the higher the silica content used, the larger the surface area, so the more effective it is to absorb methylene blue (20). However, on membranes with TEOS concentrations of 1,0 ml and 1,2 ml, there was an increase in the absorbance value due to the unbalanced and uneven matrix and filler content during the manufacture of the membrane, which caused the membrane to be less than optimal in the filtering process.

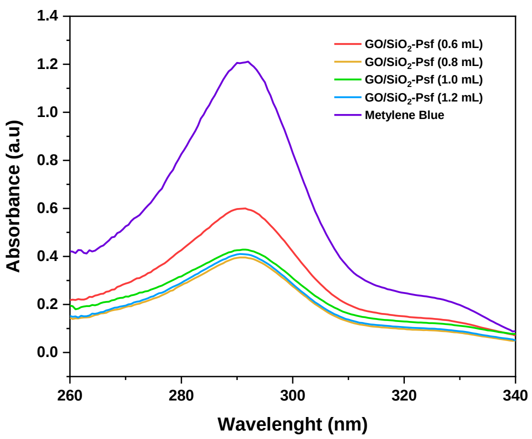

Figure 8. UV-Vis test of Methylene Blue solution 1 times filtering

Based on Fig. 8b, the methylene blue solution that has been passed through the GO/SiO_2-Psf membrane 5 times has been filtered. There is a significant decrease in the absorbance value compared to the 1 filter (Table 3). The more the filtering process uses the GO/SiO_2-Psf membrane, the lower the methylene blue content in the solution. By the statement of Neldawati, 2013 the content of methylene blue in the solution that has been passed through the membrane decreases when the filtration test is completed (21).

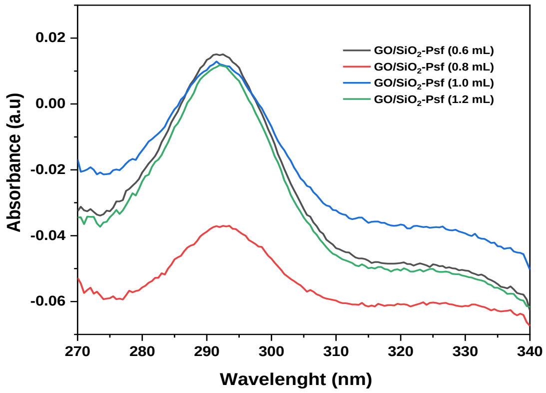

Figure 9. UV-Vis test of Methylene Blue solution 5 times filtering

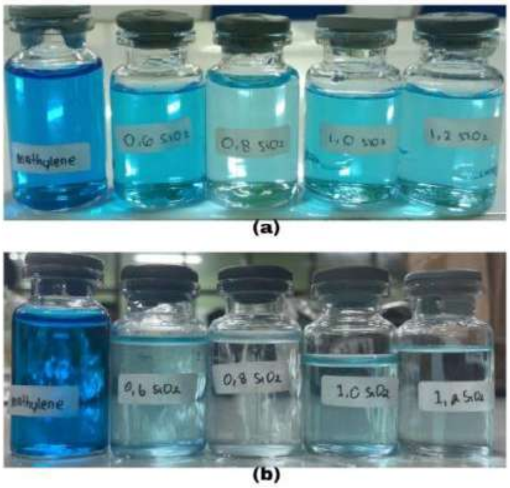

Figure 10. Metylene Blue solution filtration test results (a) 1 filter, (b) 5 filters.

Visually (Fig. 10) there is a significant color change, and it is relatively clearer when the color change is by the results of the UV-Vis test that has been carried out. The GO/SiO2-Psf membrane with TEOS 0,8 ml at 5 times of filtration obtained a negative absorbance value (Table 3), which indicates that the sample does not contain methylene blue analyte or that the methylene blue content contained in the solution is below the detection limit of the UV spectrometer method. So, it can be said that the addition of SiO_2 to the membrane can maximize the membrane as an absorbent (22), and the GO-SiO2/PSF membrane with 0,8 ml TEOS is more effective in filtering methylene blue solutions.

3.7. Flow Flux of NaCl Solution

Based on the experimental results, the data obtained are as in Table 4. The magnitude of the flow flux value is affected for different GO-SiO2/PSF membrane samples, the greater the composition of SiO^2 in the flow-flux membrane the greater. The value of the flow flux for NaCl solution and methylene blue water can be seen in Table 4. A low flux value indicates a low membrane permeability, so it can be said that the membrane is more optimal in filtering the solution, the resulting water quality is cleaner (from impurities), especially salt molecules. and methylene-blue molecules (natural dyes).

Table 3. Comparison of absorbance values of Methylene Blue solution one-times filtering and fives-times filtering

Wt% *Membrane*	*Absorbance (a.u)*	
	1 times filtering	*5 times filtering*
GO/SiO_2 *(0,6)*	*0,599*	*0,015*
GO/SiO_2 *(0,8)*	*0,396*	*-0,037*
GO/SiO_2 *(1,0)*	*0,428*	*0,012*
GO/SiO_2 *(1,2)*	*0,409*	*0,011*

The filtering time of the solution greatly affects the value of the resulting water flux, where the longer the filtering time, the GO/SiO_2-Psf membrane has a tight pore size. The GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml had the lowest flux value, namely 47,46 $L.m^{-2}.h^{-1}$ in NaCl solution and 51,55 $L.m^{-2}.h^{-1}$ in methylene blue ocean. The decrease in flux in the membrane can be caused by the closure of several pores in the membrane. The membrane flux will decrease as the filtering time increases (17-19, 48). The GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml has a tight pore size so that it is more efficient for filtering NaCl and methylene blue solutions.

Table 4. The results of the calculation of flow flux

Wt% *Membrane*	*Time (Hour)*		*Flux (L.m*$^{-2}$*.h*$^{-1}$*)*	
	NaCl (sea-water synthetic)	*Metylene Blue*	*NaCl (sea-water synthetic)*	*Metylene Blue*
GO/SiO_2 *(0,6)*	0,13	0,12	79,37	81,30
GO/SiO_2 *(0,8)*	0,17	0,16	47,46	51,55
GO/SiO_2 *(1,0)*	0,15	0,14	53,71	59,88
GO/SiO_2 *(1,2)*	0,16	0,15	48,88	54,05

3.8. Flow Flux of NaCl Solution and Methylene-Blue in Water

Fig. 11(a) is the result of one filter where the methylene blue solution is still concentrated. Fig. 11(b) is the result of 5 times filtering, where the methylene blue solution is relatively clearer. Visually, it can be seen that the GO/SiO_2-Psf membrane can reduce or absorb methylene blue gradually. Based on the results of the filtration test on the methylene blue solution 5 times filtering, the GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml looked the brightest, it was by the flux test that had been carried out.

The filtering time of the solution greatly affects the value of the resulting water flux, where the longer the filtering time, the GO/SiO_2-Psf membrane has a tight pore size. The GO/SiO_2-Psf (0.8 ml) membrane had the lowest flux value, namely 47,46 $L.m^{-2}.h^{-1}$ in NaCl solution and 51,55 L.m-2.h-1 in methylene blue ocean. The decrease in flux in the membrane can be caused by the closure of several pores in the membrane. The membrane flux will decrease as the filtering time increases (49). The GO/SiO_2-Psf (0.8 ml) membrane has a tight pore size so that it is more efficient for filtering NaCl and methylene blue solutions.

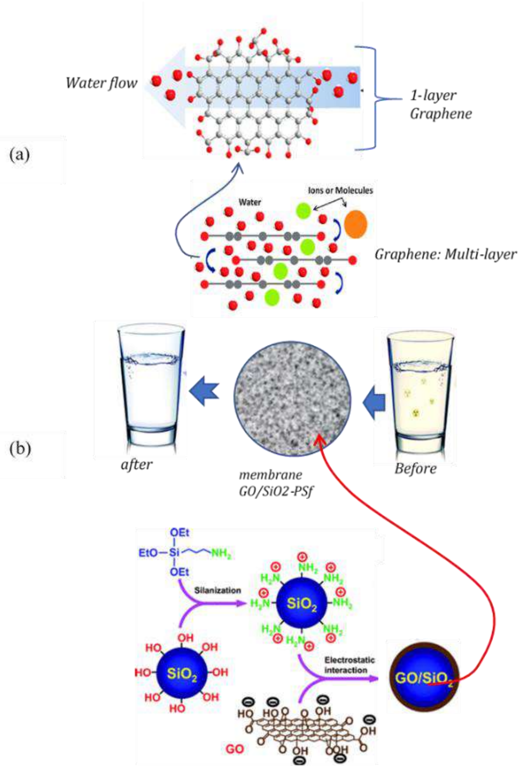

Figure 11. Filtration model of GO/SiO_2-Psf membrane: (a) water-flow in layer graphene, (b) fresh-water after filtrations.

In addition, the graphene structure that forms thin sheets of net (the order of nanometers) is very effective for filtering water molecules. GO modified with SiO2 in the membrane system is believed to be more effective in the filtration and absorption of pollutants in water (Fig. 11). Graphene oxide (GO) has unique characteristics that make it an excellent material for water purification applications. Chemically stable in water, provides high water permeability through its 2D nanochannels, and has excellent antifouling and antibacterial properties (50).

Graphene has unique physicochemical properties, extraordinarily high surface area, mechanical resistance, atomic thickness, nano-sized pores, and polar reactivity of polar and non-polar water, thus providing high selectivity and water permeability and thus providing excellent water purification efficiency. Graphene material has great potential as a membrane for water desalination, GO for good adsorption, and photocatalysis of water pollutants. On the other hand, SiO2 nanoparticles have good adsorption properties due to their high surface area and porosity, so they can be promoted as membrane fillers (47, 50-53).

Synthetic polymers, such as polysulfone, have been successfully used in water and wastewater treatment due to their unique chemical and physical characteristics, such as high chemical resistance, mechanical properties, and thermal stability. These polymeric materials tend to be hydrophobic by default and are prone to organic fouling, which requires doping with hydrophilic monomers/materials to induce hydrophilic properties to overcome fouling problems. Low-energy plasma and irradiation are used, and hydrophilic organic and inorganic materials can be mixed in the polymer by irradiation (8,50).

4. Conclusions

In conclusion, we have successfully developed GO membranes by substituting SiO2 nanoparticles in GO/SiO2 nanocomposite formations. This membrane has flexible properties and hydrophilicity for separating dyes and rejecting salt in water. The incorporation of SiO nanoparticles between the GO layers expands the vertical interlayer nano-channels, increasing the water permeability. The suitable anti-bacterial property further strengthens this membrane's application for healthy water filtration consumption. In this study, excellent results were obtained, where the Flow Flux of NaCl Solution and Methylene-Blue in Water was 79.37% and 81.30%, respectively, with a salt-rejection rate of 67.22%. However, the results of this study can still be developed further to be applied as a membrane in ultrafiltration systems.

Acknowledgments

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Manuscript Needs Major Revision (#IJE-2212-6458 (R2))

R2

Graphene Based Membrane Modified Silica Nanoparticles (GO/SiO_2-Psf) for Seawater Desalination and Wastewater Treatment: Salt Rejection and Dyes:

Manuscript ID: IJE-2212-6458

#2Reviewer1: major revision

No	Referee's Comments	Responses (for author)
1	The manuscript submitted deals with membrane synthesis and application for wastewater treatments. Authors should revise the manuscript before a concrete judgement could be suggested on the publish-ability of the work submitted to IJE.	Thank you, we noticed, and we will do it soon.
2	You should up load your highlighted revised paper.	We have highlighted the parts that have been revised (green and blue letters). We have adjusted the citations and highlighted them in green letters. I also marked the list of references, and the writing has been adjusted according to the template in the IJE journal.
3	Your response to reviewers comment should be in English.	OK, thanks you. We have fixed the previous error (R1).
4	Novelty: check of manuscript. What is the novelty of the work?	**Novelty: *check of manuscript*** - Previous studies on graphene membranes can be applied as seawater desalination membranes. - Based on previous research, SiO_2 particles can be applied to absorb dyes in water (such as methylene blue); besides that, SiO_2 nanoparticles also have antibacterial and hydrophilicity. - In this study, the Graphene/SiO_2 composite was substituted on a polysulfone polymer to form a membrane. Of course, the hope is that it will improve membrane performance in desalination and dye adsorption to obtain decent water quality, healthy for consumption.

	Explain it in the last paragraph of the Introduction section	**The last paragraph:** Previous studies on graphene membranes have shown good performance as seawater desalination membranes (9). Furthermore, silica nanoparticles can be applied as adsorption of dyes in water (such as methylene blue); In addition, SiO2 nanoparticles also have good antibacterial and hydrophilic properties. In this study, substituted graphene silica nanoparticles and polysulfone polymers were fabricated into membranes using the phase inversion method. This study is expected to improve the performance of membranes in desalination and adsorption of dyes to obtain decent water quality, healthy for consumption. GO/SiO2-Psf membrane performance in salt rejection (for desalination) and dye filtration in water (MB) will be presented in this report.
5	Novelty: check of manuscript : -penelitian sebelumnya membrane grafen dapat diaplikasi sebagai mebrane desalinasi air laut - Panda penelitian sebelumnya partikel SiO2 dapat diaplikasikan untuk absorbsi zat warna dalam air (seperti methylene blue), selain itu nanopartikel SiO2 juga bersifat antibakteri dan hydrophilicity. - Pada penelitian ini komposit Graphene/SiO2 disubtitusi pada polimer polysulfone membetuk membrane. Yang tentu harapannya akan meningkatkan performa membrane dalam hal desalinasi dan dyes adsorption, sebagai upaya untuk mendapatkan kualitas air yang layak, sehat untuk dikonsumsi.	Thanks. I'm sorry, - Previous studies on graphene membranes can be applied as seawater desalination membranes. - Based on previous research, SiO_2 particles can be applied to absorb dyes in water (such as methylene blue); besides that, SiO_2 nanoparticles also have antibacterial and hydrophilicity. - In this study, the Graphene/SiO_2 composite was substituted on a polysulfone polymer to form a membrane. Of course, the hope is that it will improve membrane performance in desalination and dye adsorption to obtain decent water quality, healthy for consumption.

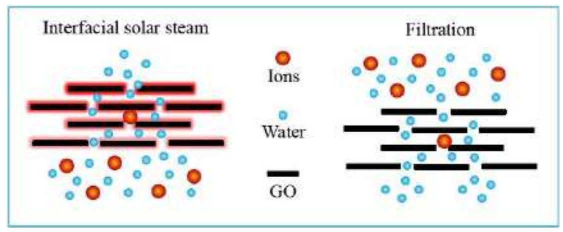
#3Reviewer2: minor revision

No	Referee's Comments	Responses (for author)
1	The manuscript submitted deals with membrane synthesis and application for wastewater treatments. Authors should revise the manuscript before a concrete judgement could be suggested on the publish-ability of the work submitted to IJE.	Thank you, we noticed, and we will do it soon.
2	What is the novelty of the work? explain it in the last paragraph of the Introduction section	Previous studies on graphene membranes have shown good performance as seawater desalination membranes (9). Furthermore, silica nanoparticles can be applied as adsorption of dyes in water (such as methylene blue); In addition, SiO_2 nanoparticles also have good antibacterial and hydrophilic properties. In this study, substituted graphene silica nanoparticles and polysulfone polymers were fabricated into membranes using the phase inversion method. This study is expected to improve the performance of membranes in desalination and adsorption of dyes to obtain decent water quality, healthy for consumption. GO/SiO_2-Psf membrane performance in salt rejection (for desalination) and dye filtration in water (MB) will be presented in this report.
3	What are PSF and TEOS. expand them in Nomenclature or in abstract	**Thank you. the terms "TEOS" and "SPF" have been expanded in the nomenclature section:** PSF = **Polysulfones** are a family of high performance thermoplastics. These polymers are known for their toughness and stability at high temperatures TEOS= Tetraethyl orthosilicate (***$Si(OH)_4$***), as a precursor to silicon dioxide, silica nanoparticles.
4	Why authors have used methylene blue?	**Methylene blue (C16H18N3SCl) is one of the most commonly used dye** stuff for dying wool, cotton and silk. Since dye-containing effluents can cause serious environmental problems, they need to be

		treated before being discharged into the surroundings. The treatment processes for removal of dye from water and wastewater can be grouped into three categories: biological (i.e. using microorganisms like bacteria, yeasts, algae and fungi), chemical (i.e. coagulation, precipitation-flocculation, oxidation, electrochemical processes), and ***physical (i.e. membrane-filtration, adsorption) [N. S. Tabrizi, 2016]*** For the dye color in the water adsorption test, several types of dyes (synthetic or natural) can be used; however, methylene blue is used to facilitate ***identification by UV-Visible spectroscopy***.
5	What is the necessity of the antimicrobial test for the membrane? did authors tested this test when the membrane was in actual work when a wastewater was used. The results may be different for independent test and in application.	The reason is that this membrane will be used as a medium for the filtration of contaminated water or desalination so that that water can be consumed healthily. So, one must also be ensured that the membrane material (Graphen/SiO_2) has good anti-bacterial properties. Graphene/SiO_2 material testing is carried out before use (in actual work). It is intended for initial performance tests before technical application and after wastewater filtration tests. Agree that the results may vary for independent testing and after application.

#4Reviewer3: minor revision

No	Referee's Comments	Responses (for author)
1	From the title, it is not easy to understand the content of the study very well. So it is suggested to change it	Previous title: ***Graphene/SiO2-Psf Membrane for Water-Filtration: Sea-Water (NaCl) and Dyes Methylene Blue (MB)*** New title: ***Graphene Based Membrane Modified Silica Nanoparticles (GO/SiO2-Psf) for Seawater and Wastewater Treatment: Salt Rejection and Dyes***
2	In the abstract; concentrate on your findings and achievements. If the authors are discussing their study should make it specific and clear and more detailed.	**New abstract**: The clean water crisis in Indonesia is increasing every year, and waste from the textile industry sector can also add to this problem. There are various water treatment processes to deal with the clean water crisis, one of which is the desalination process using graphene oxide. With the addition of hydrophilic nanoparticles, graphene oxide (GO) membranes can increase roughness and have good mechanical strength. SiO2 nanoparticles also have a high specific surface to absorb water or are hydrophilic. This study aims to determine the ability of the GO-SiO2/Psf membrane to reject salt (NaCl solution) and filtering of methylene blue solutions. Membrane prepared by variations of TEOS 0.6; 0.8; 1.0; and 1.2 ml for GO/SiO2 composite synthesis. The results showed that the GO-SiO2/PSF membrane could absorb methylene blue solution. The most optimum absorption value occurred at a TEOS concentration of 0.8 ml and had the most effective salt rejection value for NaCl solution equal to 67.22%.
3	The used methods, investigated parameters, characterization methods and techniques are all common ones, and one can hardly see any novelty in them. Can you mention the novelty of this study and advantages over other methods?	Check in manuscript, **Material and Method:** Synthesis of Graphene, Silica Nanoparticles (SiO2), or GO/SiO2 composites, as well as Graphene (GO) membranes are commonly carried out by previous researchers. The novelty is that GO/SiO2 becomes the basis of nanocomposite as a membrane filler (polysulfone, which is thermoplastic), resulting in a graphene-based membrane with

		modification of silica nanoparticles. Nanoparticles fill between GO sheets. The basic idea is that filtered water passes between the GO sheets, and SiO2 particles which are inert, porous and anti-bacterial are positioned between the GO sheets. Thus, in addition to getting pure water (H2O) with very little impurities carried, it can also ensure that it is clean from micro-organisms such as bacteria and viruses. And the final product is water that is ready, very suitable and healthy for drinking water. **The Illustrations [Xiuqiang, 2019]:**  *Notes:* ***1. The d-spacing of nanochannels (GO) is about 0.76 nm, which is larger than water molecules (0.275 nm).*** ***2. The ions (Li^+ (0.764 nm), Na^+ (0.716 nm), K^+ (0.662 nm), Mg^{2+} (0.856 nm), Ca^{2+} (0.824 nm), F^- (0. 704 nm), Cl^- (0.664 nm), Br^- (0.660 nm)) can be separated from seawater through size exclusion and electrostatic interactions, thus achieving desalination GO film filtration***
4	Page 2, column 2, line 3 correct the spelling of Graphene.	Page 2, column 2, line 3 correct the spelling of Graphene. ***Check on manuscript***
5	Section 2-3-4 what does " Cf dan Cp berturut-turut adalah konsentrasi" mean?	Section 2-3-4: Cf is the mean salt concentration in feed stream, and Cp is the salt concentration in the permeate.
6	Section 2-3-4 define the parameters of the equations.	**Section 2-3-4:** Cf is the mean salt concentration in feed stream, and Cp is the salt concentration in the permeate.
7	Provide references for peaks appeared in XRD patterns.	Reference: peak position (hkl) in XRD data, Figure2: ***Check on manuscript***

8	The figures are not numbered correctly in the text.	Thanks, I corrected
9	Page 8, column 2, the paragraph "there are 3 derivatives ...." should be moved to the introduction part.	Page 8, column 2, the paragraph "there are 3 derivatives ...." should be moved to the introduction part→**Check in manuscript (in introduction)**
10	For how long can the membrane be used optimally?	In this initial test, the filtration test with water NaCl and MB for a single-layer membrane arrangement, yet to be in a spinning membrane such as technical applications. Single sheets have shown good functional trends. So, with the technical application, it will be much more effective. Thus, the membrane can function appropriately for a standard time.

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Graphene Based Membrane Modified Silica Nanoparticles (GO/SiO2-Psf) for Seawater Desalination and Wastewater Treatment: Salt Rejection and Dyes

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ABSTRACT

The clean water crisis in Indonesia is increasing every year, and waste from the textile industry sector can also add to this problem. There are various water treatment processes to deal with the clean water crisis, one of which is the desalination process using graphene oxide. With the addition of hydrophilic nanoparticles, graphene oxide (GO) membranes can increase roughness and have good mechanical strength. SiO2 nanoparticles also have a high specific surface to absorb water or are hydrophilic. This study aims to determine the ability of the GO-SiO2/Psf membrane to reject salt (NaCl solution) and filtering of methylene blue solutions. Membrane prepared by variations of TEOS 0.6; 0.8; 1.0; and 1.2 ml for GO/SiO2 composite synthesis. The results showed that the GO-SiO2/PSF membrane could absorb methylene blue solution. The most optimum absorption value occurred at a TEOS concentration of 0.8 ml and had the most effective salt rejection value for NaCl solution equal to 67.22%.

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NOMENCLATURE			
C_f	The salt concentrations in feed	*A*	Area (m^2)
C_P	The salt concentrations in permeate	*SR*	Salt-Rejection (%)
J_w	The permeate water flux ($L\ m^{-2}\ h^{-1}$)	*F*	Flux flow
V	Volume (m^3)	A	The water permeability coefficient ($L\ m^{-2}\ h^{-1}\ bar^{-1}$)
TEOS	Tetraethyl orthosilicate, $Si(OH)_4$	Psf	Polysulfones , performance thermoplastics

1. INTRODUCTION

Indonesia is the largest archipelagic country in the world with a sea area of 5,8 million km^2 and only 2,01 million km^2 of land area. By having an existing sea area, Indonesia has great potential to be able to utilize and treat sea water as an alternative to meet the needs of clean water. The need for clean water in Java from 1991 to 2020 was 79,41 billion m^3 and is predicted to increase by 7,24 billion m^3 from 2021 to 2050, while the availability of clean water in Java is in the range from 1991 to 2020 and 2021 to 2020. By 2050, it will decrease by 75,68 billion m^3 [1].

The scarcity of clean water is caused by several factors, including the amount of water demand that continues to increase and exceeds its supply capacity [2], as well as the result of irresponsible human activities [3,4]. The scarcity of clean water also has an impact on the lower middle-class community because they have to provide funds to meet the needs of clean water. Global water scarcity is driven by water quantity and water quality issues, and measures expansion in clean water

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technologies (i.e. desalination and reuse of treated wastewater) “to reduce the number of people suffering from water scarcity” as urgently needed by the United Nations Sustainable Development Goal (SDGs 6.0) [5].

Meanwhile, the textile industry sector has increased every year. The textile industry can produce approximately 700 tons of dyes per year globally [6]. Methylene blue is one of the dyes used in the textile industry as a basic ingredient in production. Methylene blue is a heterocyclic aromatic compound that is often used in the textile, silk, and wool industries [7]. This can lead to the release of dye waste that endangers the surrounding environment and can damage aquatic organisms [8]. Based on data from the World Resources Institute (WRI), Indonesia is ranked 51st with a high-risk level of clean water crisis (high 40–80% possibility). To deal with the scarcity of clean water, there are various kinds of water treatment processes, one of which is the desalination process.

Desalination is a water purification technique by separating the levels of substances from water [9,10] by reducing ions to the required level according to human needs [11]. The desalination process can be carried out using graphene oxide. Graphene oxide offers an unusually high surface area, mechanical durability, atomic thickness, nano-sized pores and reactivity to polar and non-polar water pollutants. These characteristics provide high selectivity and water permeability, and thus provide excellent water purification efficiency. It also has the ability to adsorb and photocatalyzed water pollutants, so it has great potential for filtration materials, even for seawater desalination [9].

The addition of graphene oxide (GO) to the composite can affect the magnetization value so that it has super paramagnetic properties and can be used to absorb methylene blue [12,13]. The GO membranes contain groups such as epoxide, carboxyl, and hydroxyl which can bind to water. Graphene oxide membranes also have good mechanical strength, so they are easy to fabricate and have the potential to be produced on an industrial scale [14-16]. However, pure GO membranes have a finely stacked structure and have limited improvement in membrane performance. Based on the Cassie-Wenzel theory, there is an effective method to overcome the weakness of GO membranes, namely the addition of hydrophilic nanoparticles to increase surface roughness [17].

SiO_2 nanoparticles have properties that can carry a large number of hydroxyl groups, and the concentration of hydroxyl groups is directly proportional to the specific surface area of the amorphous silica. And also have a high specific surface so that they can absorb water or are hydrophilic, and on the surface of SiO2 there are abundant siloxane groups (Si-O-Si) that can bridge oxygen atoms [18, 19]. From this characteristic, there are many advantages, such as the broad application of biomedical and biotechnological applications, agricultural applications, industrial applications, environmental applications, and water purification. In water purification, SN can reduce biological oxygen demand, perform antimicrobial strength as a filter for water-oil mixture, and filter methylene red, commonly used in the textile and paper industry, as waste disposal from a textile factory.

There are three graphene derivatives: graphite, GO, and rGO. Graphite is the primary material of carbon which is amorphous and stacked and rich in carbon and oxygen. GO is the result of graphite oxidation which increases the amount of oxygen, and some Van der Walls bonds have been released. So the thickness of the sample has been reduced. Reduced Graphene Oxide (rGO) is a graphene oxide in which the carbon atoms of graphene undergo oxidation and reduction. In the oxidation process, there are several oxygen and hydrogen atoms bonded to carbon atoms, the result of this oxidation process is called GO. While in the reduction process, some hydrogen and oxygen bonds are released from graphene oxide so that a structure almost similar to graphene is obtained.

From various previous studies, silica nanoparticles have shown great application potential in some fields, such as chemistry, biomedicine, biotechnology, agriculture, environmental improvement, and wastewater purification. With superior properties such as mesoporous structure, high surface area, adjustable pore size/diameter, morphology, biocompatibility, modifiability, anti-bacterial, as an excellent encapsulating agent for various bioactive molecules: proven safe for targeted drug delivery, and polymer hybridization ability [20].

In a study conducted by Jiawei Sun et al. using a GO-SiO_2 membrane as an oil-in-water separator, it was found that the incorporation of SiO_2 nanoparticles with GO can expand and increase water permeability with oil rejection (>99%) for various types of oil-in-water emulsions [17, 21-22]. The GO/SiO_2 hybrid composite membrane has good hydrophilicity and thermal properties, able to reject high rhodamine B dye molecules (99%), high permeation and water resistance, so it is very good to be developed as a high-performance material for water treatment [23]. SiO_2-GO/Psf hybrid membrane presents the best overall properties, including water permeation rate, protein rejection and antifouling ability [24,25]. The SiO_2-GO nanohybrid has high hydrophilicity and good dispersibility properties derived from silica nanoparticles which are densely and uniformly coated on the GO surface and serve as a space layer of GO [23,24]. The GO-SiO_2/Psf composite under UV irradiation during filtration greatly reduces the formation of fouling and produces a high flux recovery ratio, and is effective for filtration and remove organic pollutants [26].

Previous studies on graphene membranes have shown good performance as seawater desalination membranes [9]. Furthermore, silica nanoparticles can be applied as adsorption of dyes in water (such as methylene blue); In addition, SiO_2 nanoparticles also have good antibacterial and hydrophilic properties. In this study, substituted graphene silica nanoparticles and polysulfone polymers were fabricated into membranes using the phase inversion method. This study is expected to improve the performance of membranes in desalination and adsorption of dyes to obtain decent water quality, healthy for consumption. GO/SiO_2-Psf membrane performance in salt rejection (for desalination) and dye filtration in water (MB) will be presented in this report.

2. MATERIALS AND METHOD

2.1. Materials Some of the materials used include graphite powder produced from coconut shell extract, $NaNO_3$(Merck, for analysis), $KMnO_4$ (Merck, for analysis), H_2O, H_2O_2 (Merck, 30%), NH_4OH (Merck, 25%), distilled water, and TEOS which are all obtained from Edu Lab, H_2SO_4 (Merck, 95-97%) was obtained from Indofa Industry, HCl (Merck, 37%) was obtained from Mallinckrodt, NMP solvent was obtained from Sigma-Aldrich, PSF (Polysulfone) was obtained from Sigma-Aldrich (average Mw ~35,000), dan Methylene blue (M9140, Sigma-Aldrich).

2.2. Synthesis Method

2.2.1. Synthesis of Graphene (GO) The hummer method was used, in which 5 grams of graphite powder, 2,5 grams of $NaNO_3$, and 120 ml of H_2SO_4 were placed in a 500 ml beaker with an ice bath and stirred for 30 minutes to form a black solution. Then 15 grams of $KMnO_4$ were added slowly and stirred for 30 minutes at a controlled temperature of 20°C to form a purple solution, and then stirred again for 3 hours at room temperature to form a brown solution. Then 150 ml of H_2O was added and the temperature was held at 95°C. The mixture was stirred for 3 hours at a controlled temperature of 95°C-100°C to form a dark yellow solution [27]. Then 50 ml of H_2O_2 (30%) was added slowly, then washed with HCl (1M) and H_2O until the yellow color of the solution disappeared and the pH of the solution became neutral, then sonicated for 1 hour and filtered to produce a black gel. Furthermore, the black gel was dried at a temperature of 60°C for 6 hours and produced GO.

2.2.2. Synthesis of GO/SiO_2 Composite It was carried out by the TEOS hydrolysis method, where 12,5 mg of GO was mixed with 150 ml of ethanol distilled water using a ratio of 1:5 and then sonicated for 30 minutes. Then, ammonia was added until the PH of the mixture was close to 9. Then 0.6 ml to 1.2 ml of TEOS concentration were added to the solution and stirred for 30 minutes. Then it was stirred at room temperature for 24 hours, centrifuged and washed with ethanol, then dried for 12 hours at 60°C producing a GO/SiO_2 composite [26].

2.2.3. Fabrication of GO/SiO_2-Psf Membrane Using the phase inversion method, 1 gram of polysulfone was added to 5,469 ml of NMP and stirred for 3 hours. Then 0,0333 gram of GO/SiO_2 composite was added to obtain a 0.5wt% solution and sonicated for 30 minutes to shorten the dissolution process. No air bubbles were formed in it. Then the solution was formed on a glass plate and soaked in distilled water for 24 hours. Then it was dried for 24 hours at room temperature, which finally produced a GO/SiO_2-Psf membrane [28].

2.3. Characterizations

2.3.1. Characterization of GO/SiO_2 and GO/SiO_2-Psf Membrane Phase and crystal structure analysis of nanoparticles (GO, SiO_2) and GO/SiO_2 nanocomposites used X-Ray Diffraction (PANalytical X'Pert Pro) plus Expert High Score Plus software; using monochromatic radiation CuKα (40kV/40 mA) taken at an angle of $5° \leq 2\theta\_ \leq 90°$ at a rate of 0.02°/min at room temperature. Functional group analysis using infrared wave absorption test using FTIR (Shimadzu Brand, Type: IR-Prestige 21). Lattice vibration analysis in Graphene phase using Ramman spectroscopy (wavelength range 4000-200 cm^{-1}), and to observe the morphology of GO/SiO_2 nanocomposite and GO/SiO_2-Psf membrane using the Scanning electron microscope/SEM test (FEI Brand, Type: Inspect-S50). UV-Visible test to analyze the adsorption of methylene blue dye in water. And the analysis of the hydrophilicity of the membrane surface with the CAA 2320 Contact Angle instrument.

2.3.4. Filtration of NaCl Solution GO/SiO_2-Psf was placed in a funnel Buchner flask and a hose was connected to a vacuum pump (Rocker 300) (Fig. 1). The vacuum pump was turned on, then the membrane was dripped with 10 ml of NaCl solution (as sea-water synthetic) at a pressure of 650 mmHg for all variations of the GO/SiO_2-Psf membrane. The membrane that has been passed through the NaCl solution is then weighed.

2.3.5. Filtration of Methylene Blue in Aqueous The 20 ppm methylene blue solution was passed through a 12,25 cm^2 GO-SiO_2/PSF membrane placed in a Buchner flask funnel and connected to a vacuum pump (Rocker 300) using a tube (Fig.1). The vacuum pump was turned on, then the membrane was dripped with 10 ml of methylene blue solution at a pressure of 650 mmHg for all variations of the GO-SiO_2/PSF membrane. The results of the filtered methylene blue solution will be subjected to a

UV-Visible test to determine the absorbance [29]. The results of the filtered methylene blue solution will be tested by UV-Vis (Shimadzu 1800) with a wavelength of 200-600 nm, to determine the absorption.

Methylene blue (C16H18N3SCl) is one of the most commonly used dyes for dyeing wool, cotton and silk. Since sewage or water containing dyes can cause serious environmental problems and the availability of healthy water for consumption, it is necessary to treat these wastes before being discharged into the environment [30].

2.3.6. Salt-Rejection Test A salt-rejection test was carried out to determine the efficiency of the membrane in filtering NaCl solution. Salt-rejection (SR) can be obtained by the following formula [11]:

$$Salt-rejection = \frac{Cf - Cp}{Cf} x100 \qquad (1)$$

Where, Cf is the mean salt concentration in feed stream, and Cp is the salt concentration in the permeate.

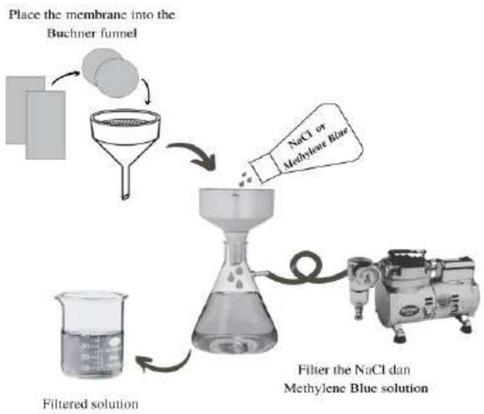

Figure 1. Filtration test with GO/SiO_2-Psf

2.3.5. Flow-Flux Test The membrane flux test was carried out to measure the optimization parameters of the membrane. The *filtrate flow flux* (J) can be obtained by the formula [11]:

$$J = \frac{V}{At} \qquad (2)$$

Where A is membrane area (m^2), V is the volume of filtrate generated (liter), and t is process time (hours).

3. RESULT AND DISCUSSION

3.1. X-Ray Diffraction of Composite Diffraction analysis of Graphene (GO) material and GO/SiO_2 composite is presented in Fig. 2. The peak at 2θ= 10.21° is associated with the (002) GO plane in the GO/SiO_2 composite, but this peak does not appear. It is associated with weak diffraction, and the presence of graphite oxide heaps, in which SiO_2 is coated with a GO sheet, causes GO diffraction to appear [22]. The similarity of the diffraction peaks for GO and the GO/SiO_2 composite is indicated at positions 2θ =21.8° and 26.2°. And the presence of a widened peak at 2θ = 23.31° indicates that silica is amorphous [29].

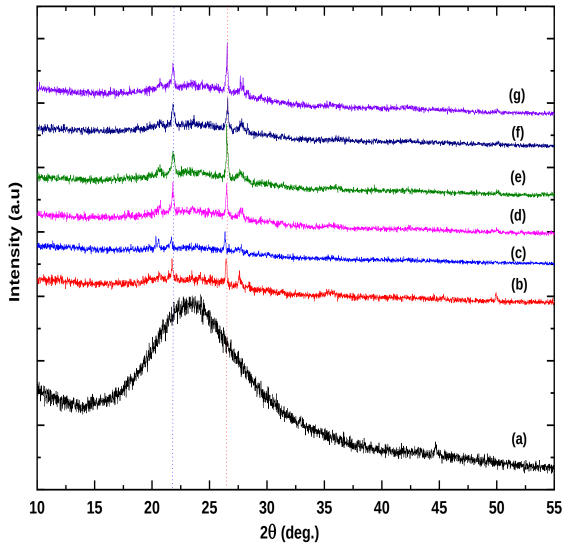

Figure 2. Pattern Diffraction of SiO_2(a), Graphite(b), GO (c) and GO/SiO_2-Psf Composite (d-g)

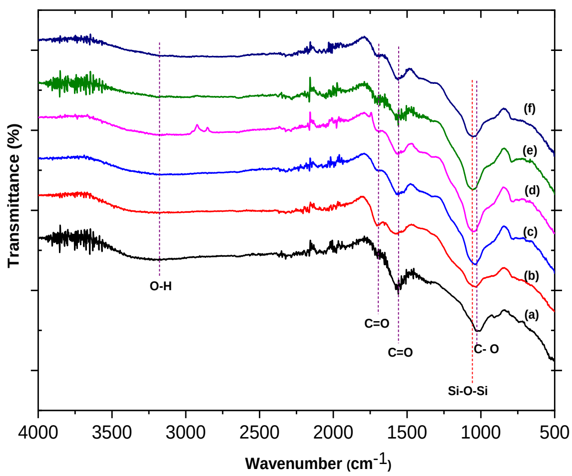

Figure 3. Functional group of GO and GO/SiO_2-Psf: (a) Grafite, (b) GO, dan (c-f) composite.

3.2. Functional Group of Composite The functional group analysis of the characteristics for GO materials and GO/SiO_2 composites is presented in Fig. 3. The results of the analysis showed the presence of silanol groups and Si-O silicates and C-H carbon groups. These functional groups are a representation of the composite material [13,32]. The GO/SiO_2 composite exhibits a new peak at 1100 cm^{-1}, characteristic of the Si-O-Si asymmetric vibration. The presence of this absorption proves that silica is on the GO surface [22]. In addition, the peak at 1600-1735 cm^{-1} is associated with the C=O vibration of the carboxylate group. The decreasing C=O absorption peak in the composite was due to the interaction of C=O with GO to become Si-O-C indicating a bonding interaction between GO and Silica.

3.3. Morphology of GO, GO/SiO_2 Composite and GO/SiO_2-Psf Membrane The morphology of GO/SiO_2 composite material and GO/SiO_2-Psf membrane with NPM solvent during the membrane preparation process is presented in Fig. 4. The shape of the particles varies, it appears that the particles have several (small) pores, and between the stacks of particle arrangement there are quite large voids. Particle sizes also vary, small and large, this affects the formation of voids which will later play a role in forming the density of the membrane material.

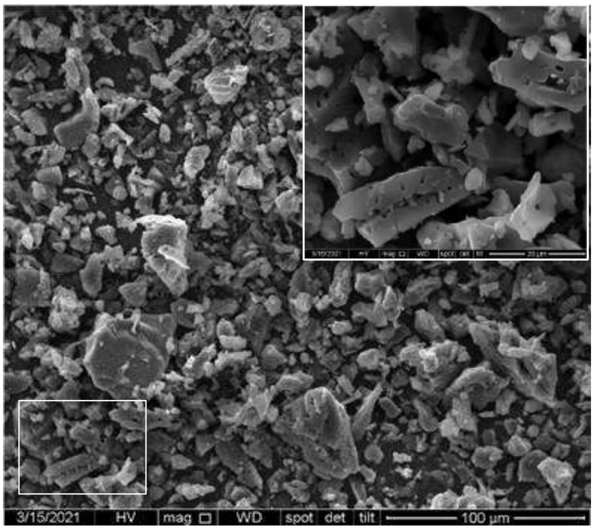

Figure 4. Morphology of GO/SiO_2 composite

The morphology of the GO/SiO_2-Psf membrane material prepared by the phase inversion method is presented in Fig. 5. It can be seen on the surface of the membrane and its cross section. On the surface of the membrane, pores of varying sizes were seen, and these were identified as water inlet channels through the membrane through the cavities formed on the inside of the membrane. The size and shape of the cavity is influenced by the type of membrane polymer (polysulfone) and the composite material embedded in the membrane. Figure 5b shows a cross-sectional view of the modified composite membrane with the topical asymmetrical morphology of the membrane fibers. The middle layer represents the predominant morphology with finger-like structures. This finger-like structure is characteristic of an asymmetrical membrane where the cross-section of the membrane consists of a finger structure with a porous underlayer [32]. The formation of a porous surface on GO-SiO_2-psf is caused by an increase in the hydrophilic nature of the solution, which will accelerate the rate of solvent exchange [33]. The addition of GO into the membrane causes a larger cavity. In theory, the more macro-voids appear, the greater the membrane's permeability [32].

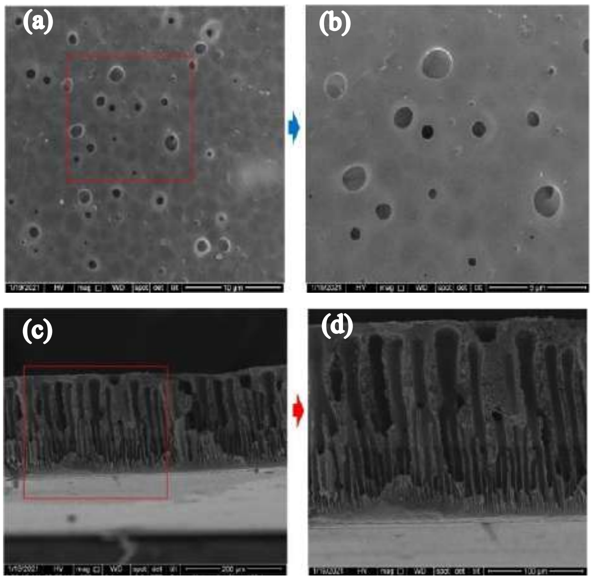

Figure 5. Morphology of GO/SiO_2-Psf membrane: (a-b) *surface pores*, (c-d) *cross-sectional view*.

3.4. Hydrophobicity of GO/SiO_2-Psf Membrane The hydrophilic nature of the membrane has an essential role in filtration performance. In principle, the hydrophilicity of the membrane can be determined by the water contact angle.

The contact angle of GO/SiO_2-Psf polysulfone membrane with NMP solvent is presented as follows, for the weight percent of GO/SiO_2 composite (for SiO_2, *Wt%* ≈ 0.5%) with the percentage of composite material varied: 0.6; 0.8; 1.0; and 1.2%. Fig.5 and Table 1 show that the angle is smaller than 90° (hydrophobic): each membrane sample shows an average contact angle of: 75.41°; 70.16°; 76.78°; and 74.25°. The lower the contact angle, the higher the membrane hydrophilicity [34; 35]. The greater contact angle value is caused by the surface tension between the membrane and the water. On the other hand, for the SiO_2-Psf membrane contact angle (without GO) the contact angle is 71.13°, approximately the same as the GO/SiO_2-Psf membrane. This indicates that the membrane is hydrophilic or slightly water-loving (contact angle (θ) ($0° \le \theta \le 90°$) [36,37]. Furthermore, a small contact angle generally results in better hydrophilicity, increased water flux, and resistance to impurities [38]. Thus, the experimental results obtained in this study are in accordance with the literature [39].

3.5. Anti-Bacterial of GO/SiO_2 Composite The disc diffusion test was carried out to determine the inhibitory power of the compound on bacterial growth. Bacterial suspension (∅D600 nm 0.1) was rubbed on the surface of Muller Hinton Agar (MHA) media on a Petri dish using a sterile cotton swab. A paper disk containing 20 *l* of the test compound was placed on the surface of the MHA. Incubation was carried out for 48 hours at 30ºC. The clear zone formed around the disc was expressed as the inhibitory power of the compound against bacterial growth. Anti-bacterial test in this study, using 2 types of bacteria, namely Escherichia coli and Staphylococcus aureus.

The results of the antimicrobial test of GO/SiO_2 against Escherichia coli and Staphylococcus aureus with the disc diffusion method (Table 2) showed that the inhibitory power of E. Coli was stronger than S. Aureus; The increase in GO-SiO2 (ppm) concentration further strengthens the antibacterial activity of the E. Coli strain, compared to S. aureus which tends to remain stable. Thus, the membrane material will be confirmed to have resistance to bacterial growth (E. Coli and S. Aureus) [37]. Bacterial-mediated infections can cause various acute or chronic diseases and antibiotic resistance in pathogenic bacteria has become a serious health problem. Graphene-based materials have been very well studied due to their outstanding bactericidal activity on various bacteria. The use of GO material in membrane preparation will provide biosafety advantages [24]. Likewise, the presence of SiO_2 nanoparticles, also has excellent properties as an anti-bacterial material [40,41].

3.6. Salt-Rejection of NaCl Solution Based on Table 4, the results of the salt-rejection calculation, the Cp value is obtained from the total solid NaCl dissolved in freshwater with a salt density of 2,16 g/ml, and the Cf value is obtained from the initial concentration in the NaCl solution, which is 27,79 g/ml. The GO/SiO_2-Psf membrane with a TEOS (0,8 ml) showed the most effective results for seawater desalination because the GO/SiO_2-Psf membrane was able to filter out NaCl compounds in solution by 67,22%. The high salt-rejection value is caused by the distribution of silica grains on the membrane being very tight so that the pores are getting smaller and able to filter NaCl compounds efficiently [15]. The research of Zeng et al. (2019) showed that an increase in membrane thickness due to the pore density of SiO_2, can help reduce conductive losses in bulk water and increase salt rejection ability, but energy efficiency is limited by maximum liquid flux [42]. The GO/SiO_2-Psf membrane with TEOS concentrations of 1,0 ml and 1,2 ml decreased because the ratio of matrix and filler was not balance.

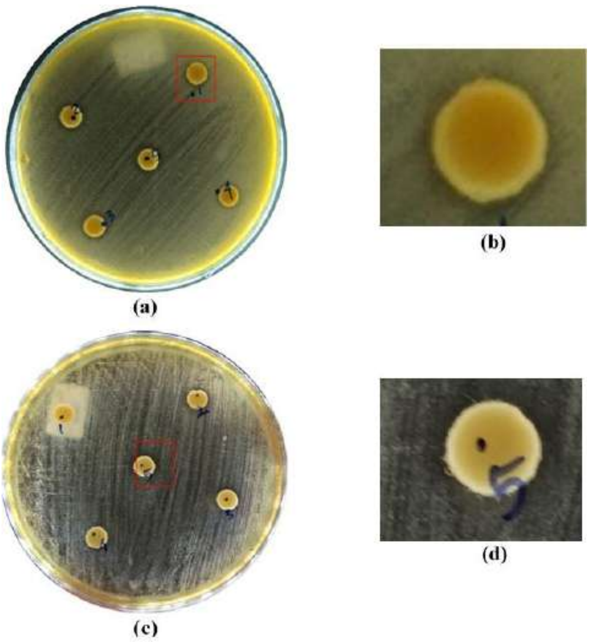

Figure 6. Photograph of antimicrobial of GO/SiO_2: (a-b) E. Coli and (c-d) S. Aureus

TABLE 1. Contact Angle of GO/SiO_2-Psf(0.5%) Membrane with DMAC and NMP Solvents

PSF-0,5% GO/SiO_2	*Contact Angle (degrees) with DMAC solvent*			*Contact Angle (degrees) with NMP solvent*		
	Right	*Left*	*Average*	*Right*	*Left*	*Average*
0,60 ml TEOS	76,13	81,02	***78,58***	74,39	76,44	***75,41***
0,80 ml TEOS	61,78	60,36	***61,07***	69,71	70,61	***70,16***
1,00 ml TEOS	82,71	82,53	***82,62***	72,65	80,90	***76,78***
1,20 ml TEOS	70,24	70,86	***70,55***	74,79	73,72	***74,25***
0,5% SiO_2-PSF	69,91	72,44	***71,18***	70,43	71,84	***71,13***

TABLE 2. Antimicrobial of GO-SiO_2 compounds against E. coli and S. Aureus

Bacterial	*test repeats*	*Inhibition zone diameter (cm) at various concentrations of GO-SiO_2 compounds*				
		100 ppm	200 ppm	300 ppm	400 ppm	500 ppm
Escherichia coli	1	0,83	0,94	1,01	1,08	0,97
	2	0,79	0,83	0,91	0,95	1,04
	3	0,93	0,94	0,94	0,98	1,01
Staphylococcus aureus	1	0,55	0,55	0,55	0,55	0,55
	2	0,55	0,55	0,55	0,55	0,55
	3	0,55	0,55	0,55	0,55	0,55

3.6. Filtration of Methylene Blue in Aqueous By using 20 ppm methylene blue was then carried out a filtration test. The methylene blue solution was filtered using a GO/SiO_2-Psf membrane with various concentrations of TEOS 0,6; 0,8; 1,0; and 1,2 ml. The results of the methylene blue solution filtration test (Fig.2) show that the methylene blue solution, which has been filtered using a GO/SiO_2-Psf membrane, appears to change in color before and after filtering.

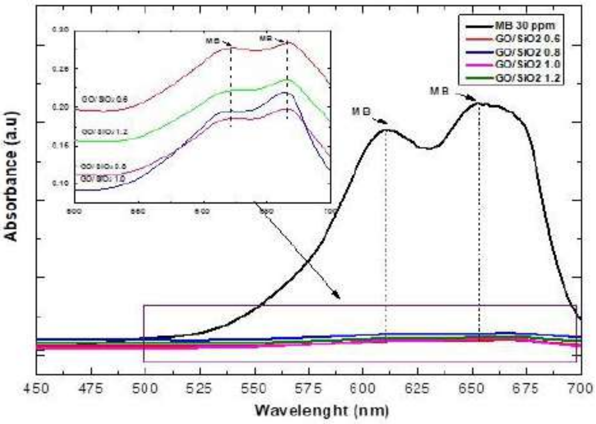

Figure 7. Dye absorption in water (Methylene Blue) by GO/SiO_2 nanocomposite.

The UV-Vis test was carried out using a wavelength of 200 to 600 nm, and the absorption value of each sample was obtained (Fig.3). The methylene blue solution without passing through the GO-SiO_2/Psf membrane obtained an absorbance value of 1,206 at a wavelength of 291 nm. Figure 7 shows the results of the UV-Vis test showing the relationship between wavelength and absorbance. The UV-Vis absorption spectrum of GO/SiO_2 nanocomposite has a strong absorption in the range of 200-650 nm. In the composite for TEOS (0.6 ml), GO showed a characteristic peak at 325 nm, for TEOS (0.8 ml) it decreased starting at the peak of 280 nm. For TEOS (1.0 and 1.2 ml) indicates a stable position and continues to increase. The maximum absorption peak corresponds to the π-π* transition bond in the C=C aromatic structure and the $\pi \rightarrow \pi$* transition in the carbonyl group (C=O). The characteristic peak shifts to 270 nm for GO due to the presence of reduced graphene [21,45].

The Methylene blue, which was passed through the GO/SiO_2-Psf membrane in one time filtering (Fig. 8a) decreased the absorption value, where the most optimum absorbance occurred in the membrane with a TEOS concentration of 0,8 ml, which was 0,396 at a wavelength of 291 nm. This is to the literature conducted by Nurul et al, 2019 where the absorbance of methylene blue can be carried out using silica [17]. This also supports the statement of Cahyo et al, 2018 that the higher the silica content used, the larger the surface area, so the more effective it is to absorb methylene blue [20]. However, on membranes with TEOS concentrations of 1,0 ml and 1,2 ml, there was an increase in the absorbance value due to the unbalanced and uneven matrix and filler content during the manufacture of the membrane, which caused the membrane to be less than optimal in the filtering process.

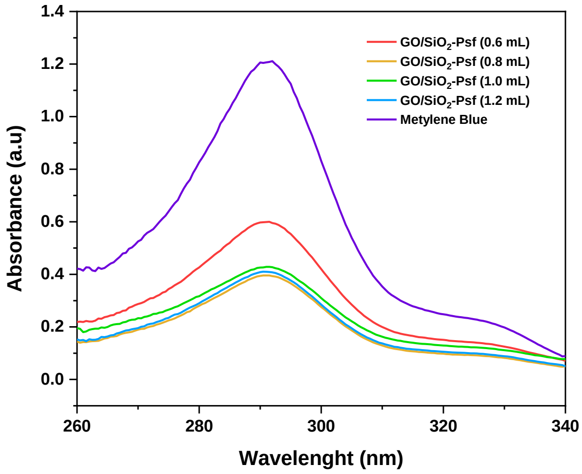

Figure 8. UV-Vis. test of Methylene Blue solution 1 times filtering

Based on Fig. 8b, the methylene blue solution that has been passed through the GO/SiO_2-Psf membrane 5 times has been filtered. There is a significant decrease in the absorbance value compared to the 1 filter (Table 3). The more the filtering process uses the GO/SiO_2-Psf membrane, the lower the methylene blue content in the solution. By the statement of Neldawati, 2013 the content of methylene blue in the solution that has been passed through the membrane decreases when the filtration test is completed [18].

Visually (Fig. 10) there is a significant color change, and it is relatively clearer when the color change is by the results of the UV-Vis test that has been carried out. The GO/SiO2-Psf membrane with TEOS 0,8 ml at 5 times of filtration obtained a negative absorbance value (Table 3), which indicates that the sample does not contain methylene blue analyte or that the methylene blue content contained in the solution is below the detection limit of the UV spectrometer method. So, it can be said that the addition of SiO2 to the membrane can maximize the membrane as an absorbent (22), and the GO-SiO2/PSF membrane with 0,8 ml TEOS is more effective in filtering methylene blue solutions.

TABLE 3. Comparison of absorbance values of Methylene Blue solution one-times filtering and fives-times filtering

Wt% Membrane	Absorbance (a.u)	
	1 times filtering	5 times filtering
GO/SiO_2 (0,6)	0,599	0,015
GO/SiO_2 (0,8)	0,396	-0,037
GO/SiO_2 (1,0)	0,428	0,012
GO/SiO_2 (1,2)	0,409	0,011

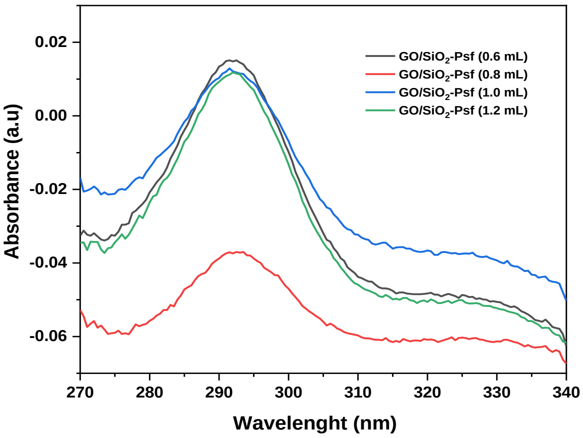

Figure 9. UV-Vis test of Methylene Blue solution 5 times filtering

3.7. Flow Flux of NaCl Solution Based on the experimental results, the data obtained are as in Table 4. The magnitude of the flow flux value is affected for different GO-SiO2/PSF membrane samples, the greater the composition of SiO^2 in the flow-flux membrane the greater. The value of the flow flux for NaCl solution and methylene blue water can be seen in Table 4. A low flux value indicates a low membrane permeability, so it can be said that the membrane is more optimal in filtering the solution, the resulting water quality is cleaner (from impurities), especially salt molecules. and methylene-blue molecules (natural dyes).

The filtering time of the solution greatly affects the value of the resulting water flux, where the longer the filtering time, the GO/SiO_2-Psf membrane has a tight pore size. The GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml had the lowest flux value, namely 47,46 $L.m^{-2}.h^{-1}$ in NaCl solution and 51,55 $L.m^{-2}.h^{-1}$ in methylene blue ocean. The decrease in flux in the membrane can be caused by the closure of several pores in the membrane. The membrane flux will decrease as the filtering time increases [17-19, 46]. The GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml has a tight pore size so that it is more efficient for filtering NaCl and methylene blue solution

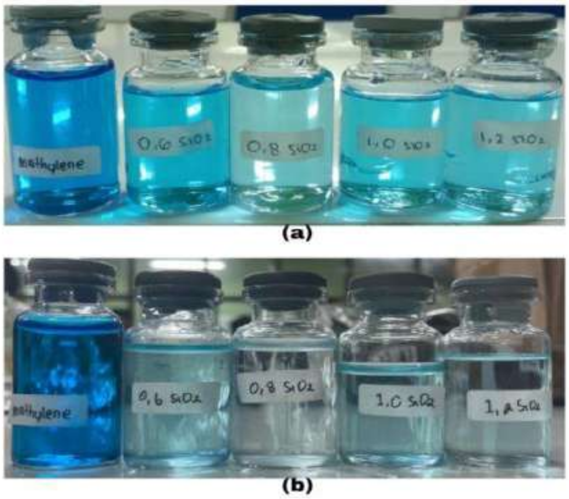

Figure 10. Metylene Blue solution filtration test results (a) 1 filter, (b) 5 filters.

TABLE 4. The results of the calculation of flow flux

Wt% Membrane	*Time (Hour)*		*Flux ($L.m^{-2}.h^{-1}$)*	
	NaCl (sea-water synthetic)	*Metylene Blue*	*NaCl (sea-water synthetic)*	*Metylene Blue*
GO/SiO_2 (0,6)	0,13	0,12	79,37	81,30
GO/SiO_2 (0,8)	0,17	0,16	47,46	51,55
GO/SiO_2 (1,0)	0,15	0,14	53,71	59,88
GO/SiO_2 (1,2)	0,16	0,15	48,88	54,05

3.8. Flow Flux of NaCl Solution and Methylene-Blue in Water Fig. 11(a) is the result of one filter where the methylene blue solution is still concentrated. Fig. 11(b) is the result of 5 times filtering, where the methylene blue solution is relatively clearer. Visually, it can be seen that the GO/SiO_2-Psf membrane can reduce or absorb methylene blue gradually. Based on the results of the filtration test on the methylene blue solution 5 times filtering, the GO/SiO_2-Psf membrane with a TEOS concentration of 0,8 ml looked the brightest, it was by the flux test that had been carried out.

The filtering time of the solution greatly affects the value of the resulting water flux, where the longer the filtering time, the GO/SiO_2-Psf membrane has a tight pore size. The GO/SiO_2-Psf (0.8 ml) membrane had the lowest flux value, namely 47,46 $L.m^{-2}.h^{-1}$ in NaCl solution and 51,55 L.m-2.h-1 in methylene blue ocean. The decrease in flux in the membrane can be caused by the closure of several pores in the membrane. The membrane flux will decrease as the filtering time increases [47]. The GO/SiO_2-Psf (0.8 ml) membrane has a tight pore size so that it is more efficient for filtering NaCl and methylene blue solutions.

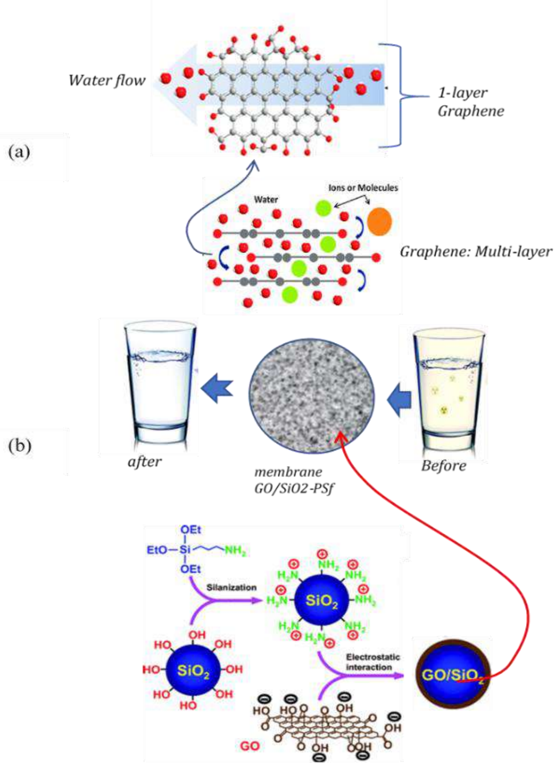

Figure 11. Filtration model of GO/SiO_2-Psf membrane: (a) water-flow in layer graphene, (b) fresh-water after filtrations.

In addition, the graphene structure that forms thin sheets of net (the order of nanometers) is very effective for filtering water molecules. GO modified with SiO2 in the membrane system is believed to be more effective in the filtration and absorption of pollutants in water (Fig. 11). Graphene oxide (GO) has unique characteristics that make it an excellent material for water purification applications. Chemically stable in water, provides high water permeability through its 2D nanochannels, and has excellent antifouling and antibacterial properties [48].

Graphene has unique physicochemical properties, extraordinarily high surface area, mechanical resistance, atomic thickness, nano-sized pores, and polar reactivity of polar and non-polar water, thus providing high selectivity and water permeability and thus providing excellent water purification efficiency. Graphene material has great potential as a membrane for water desalination, GO for good adsorption, and photocatalysis of water pollutants. On the other hand, SiO2 nanoparticles have good adsorption properties due to their high surface area and porosity, so they can be promoted as membrane fillers [45, 48-51].

Synthetic polymers, such as polysulfone, have been successfully used in water and wastewater treatment due to their unique chemical and physical characteristics, such as high chemical resistance, mechanical properties, and thermal stability. These polymeric materials tend to be hydrophobic by default and are prone to organic fouling, which requires doping with hydrophilic monomers/materials to induce hydrophilic properties to overcome fouling problems. Low-energy plasma and irradiation are used, and hydrophilic organic and inorganic materials can be mixed in the polymer by irradiation [8,48].

4. CONCLUSIONS

In conclusion, we have successfully developed GO membranes by substituting SiO2 nanoparticles in GO/SiO2 nanocomposite formations. This membrane has flexible properties and hydrophilicity for separating dyes and rejecting salt in water. The incorporation of SiO nanoparticles between the GO layers expands the vertical interlayer nano-channels, increasing the water permeability. The suitable anti-bacterial property further strengthens this membrane's application for healthy water filtration consumption. In this study, excellent results were obtained, where the Flow Flux of NaCl Solution and Methylene-Blue in Water was 79.37% and 81.30%, respectively, with a salt-rejection rate of 67.22%. However, the results of this study can still be developed further to be applied as a membrane in ultrafiltration systems.

5. ACKNOWLEDGMENTS

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