An Experiment of Hybrid Microfiltration Membranes (MFM) Using Dead-End-Flow Method on Water Source

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Abstract The focus of this experiment is to investigate advisability of hybrid microfiltration membrane against natural water qualities. Hybrid membrane microfilter (MF) was produced through casting method. Moringa olifera powder mixed with activated carbon of coconut shell charcoal powder were used as reinforcement and biopolymer ceresin and polyvinyl alcohol (PVA) used as composite matrix. In order to testing hybrid MF membrane of turbidity of water the method of Dead-End-Flow (DEF) was carried out. The main purpose is to investigate the turbidity and swelling of hybrid MF membrane after water filtration. The experiment result shows moringa oleifera as an organic has good function to reduce water pollutant like bacteria. Then, activated carbon of coconut shell charcoal own has unique characteristic in water treatment are eliminate the water color, odor, toxin absorb and chemicals. In addition, hybrid MF membrane with moringa oleifera/activated carbon coconut shell charcoal/PVA/ceresin has been impacted against to turbidity value of water sources. Then swelling of hybrid MF membrane was increased with increasing member of moringa oleifera in specimen. It can be concluded that hybrid MF membrane is an alternative for water purification.

Key words; **Hybrid, Membrane, Microfiltration, Dead end flow, Turbidity.**

I. Introduction

Since the people knowing water quality decreasing membrane has been developed widely for purification the source water and wastewater. Membrane was classified based on the porosity of filtrate including microfiltration (MF), ultra-filtration (UF) Nano-filtration (NF), and reverse osmosis (RO) [1]. Theoretically, membrane is wall that separates the two phases and controls the transportation of different chemical components in a specific manner. In addition, membrane can be defined as separator that adjusts the leakage and seepage rates of its adjacent chemical components [2]. Microfiltration is a low pressure separation process utilizing membranes with very open pore structures. Microfiltration (MF) is a type of physical filtration that has micro pore size. MF developed specifically to remove complex pollutant in water source such as microbial removal, protein fractionation, and pretreatment. The pore size used for microfiltration ranges from about 0.1 to 10 µm. In the present, microfiltration can also be applied combination with reverse osmosis, nano-filtration, and ultra-filtration. Water treatment by using membrane many advantages can be taken such as variety in application, low energy, and it can be hybrid. [3-5]. However, the disadvantage of membrane in water treatment process are easier polarization formed (fouling), less selective, and low durability [6].

Since last ten years, membrane composite increased applied on water purification process due to many kinds of pollutants. In order to improving ability of membrane on water purification and answering an environmental issue, membrane made using organic material has been developed in the present. Several experiments about membrane using organic material have been carried out. Moringa oleifera was popular used for water purification because it has ability to non-toxic, bacteriology resistant [7], and recycle color of water [8-10]. Meneghel [11] and Agnihotri [12] studied about the moringa oleifera in function for remove fluoride on the water. According to the ability of moringa oleifera on water purification process is limited. Presently, activated carbon has been used for water purification due to their ability is good in reduce water smelt and chemical absorption [13-18]. In addition, beside of both materials for water purification the polymer also can be used such polyvinyl alcohol (PVA). Haryadi et.al [19] study characteristics of composite membrane based on the PVA-TMSP *sulfonated* for direct methanol fuel cell (DMFC) application. Afterwards, the characterization of membrane using *polysaccharides*/PVA blend nanofibrous manufactured by electrospinning method has been studied [20].

Composite membranes have popular used in water purification as explained above. According to the result for each material application on the membranes, hybrid composite is a potential approach to improve abilities of membrane. Incorporation more than one type of material in membranes production is approach to improve membrane abilities in reduces pollutants on the water. The researchers that are focus in developing hybrid microfiltration membranes like Ravindran et.al [21]. In this research hybrid membrane bioreactor technology was investigated for treatment of natural waters. In addition, the biological and adsorption methods use hybrid membrane with activated carbon treatment for produced drinking water was reviewed by Stoquart et.al [22]. Enhancement membrane filtration using the hybrid anaerobic membrane bioreactor coupled with online ultrasonic equipment for waste activated of sludge was conducted by Xu. et.al [23]. According to the above information almost all membranes manufactured only used single function in purification water from the pollutants contain. The problem is how to remove all pollutant from the water in single membranes.

In the research, hybrid microfiltration membranes use two particles reinforcement and bio-polymer solution was designed for purification water. The aim is to investigate the functionality and abilities of hybrid membranes microfiltration on filtrating water pollutants. Swelling, Mechanical and physical test were carried out and membranes failure is analyzed using the SEM. The fictionalization of hybrid membrane was determined using the BOD and Dead-end-Flow (DEF).

II. Membrane Manufacture

A. Material

In manufacturing hybrid microfiltration membrane two micro-particles used such moringa Olivier (MO) powder and activated carbon coconut shell charcoal (CA). Both powder mixed based on the weight fraction ratio as noted on Table 1. Whilst, bio-polymer such polyvinyl alcohol and ceresin of cocoon were applied as a matrix (see Table 1). Fig. 1a to d show the materials of hybrid microfiltration membrane including moringa Olifera, activated carbon, PVA and Ceresin (SC).

As explained above, moringa oliefra is an organic material that has ability to degrade water color effective until 98%. Their also can degrade biochemical oxygen demand (BOD) and mud contents are 62% and 70 ml per liters, respectively. In addition, moringa can clean the Escherichia coli per liter water about 90% in 20 minutes. This ability due to of moringa oleifera structured by protein cluster consists of 79.3% cationic and 20.7% anionic. Besides it contains 4-(α-L-rhamnopyranosyloxy) benzyl isothiocy-anate are resistant to the bacteria and the protein will have positive content in water dissolved.

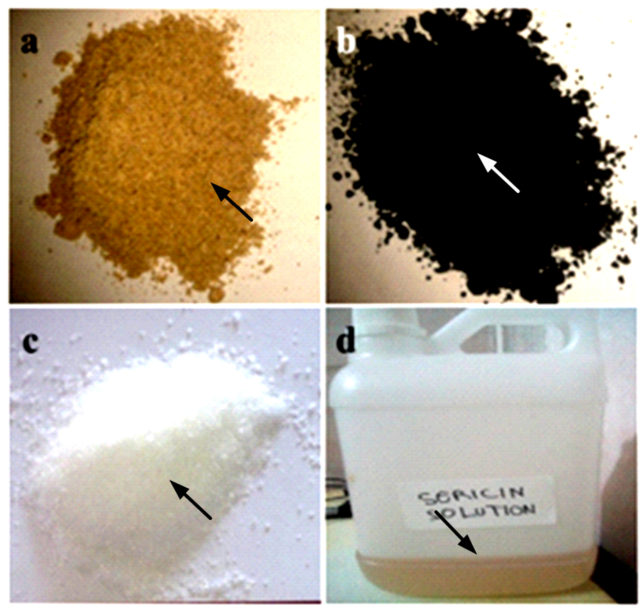

Fig. 1. a. Moringa oleifera; b. Activated carbon coconut shell charcoal; c. *Polyvinyl alcohol* (PVA); d. Cocoon ceresin

Activated carbon (AC) coconut shell charcoal other material is used as reinforcement of hybrid MF membrane. Activated carbon is a porous solids that containing 85% until 95% carbon. AC has macro pore size and it has advantage for gas adsorption, remove the smelt and color of water. The weight of AC coconut shell charcoal is 15% to 19% of the total weight of coconut. Ceresin is a substance contained in silk cocoon of 20-30%. Here, ceresin is consisting of *amino acid hydrophilic* of 70% and hydrophobic less than 20%. *Polyvinyl Alcohol* (PVA) is kind of polymers with hydrophilic behavior, elastic, corrosion resistant, and adhesiveness. In addition, PVA are not smelt, yellowish and soft. The PVA density is 1.3gr/cm^3 at 20°C with pH 3.5-7.0 on 40 gram per liters water solvent.

TABLE 1.
HYBRID MF MEMBRANES VARIATION

Membrane classification	Code	Solution Concentrations (%wt)			
		MO	CA	PVA	SC
MBH1	H1	7.5	22.5	35	35
MBH2	H2	15	15	35	35
MBH3	H3	22.5	7.5	35	35
MBH4	H4	23.1	23.1	70	0

B. Manufacture process

Hybrid MF membrane with solution bio-polymer (PVA/ceresin)/moringa particles/AC coconut shell charcoal was manufactured based the casting method. Table 1 shows the hybrid composition of MF membranes.

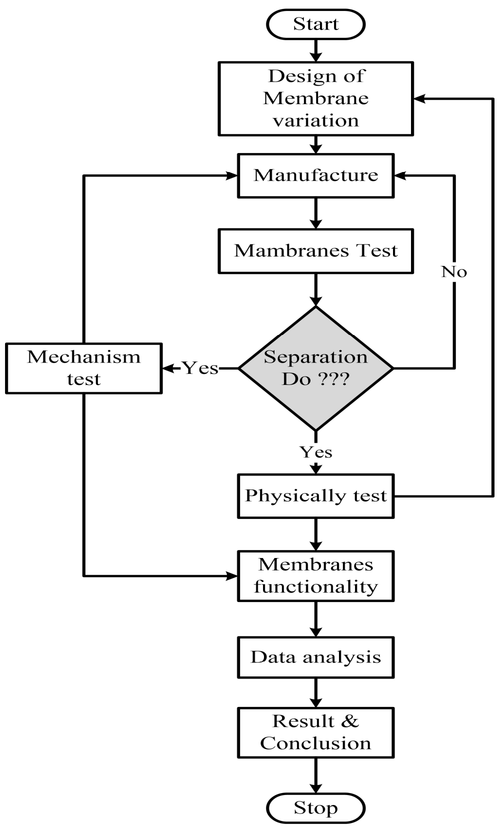

Fig. 2. Schematics of hybrid MF membranes process

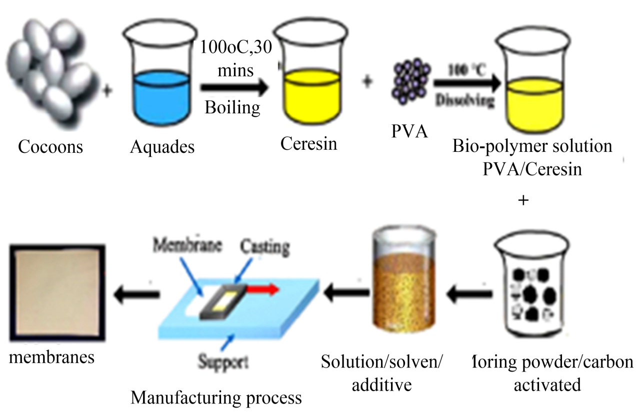

Fig. 3 Membranes manufacturing process

Fig. 3 illustrate hybrid microfiltration membranes manufacturing process. The casting method to produce membranes was conducted. The manufacture of membranes is begun by boiling the cocoon at 100oC temperature as long as 30 minutes to produce ceresin. The next step is make solution of PVA and ceresin are stirred on speeds of 250 rpm using the 20mg magnetic stirrer. In this process solvent 5% was added. Next, the moringa powder and the activated carbon coconut shell charcoal based on the Table 1 for every hybrid membranes design are mixed manually. Afterwards, bio-polymers and powder mixtures are mixed using the stirrer machine on constant speed 250 Rpm as long as 24 hours using magnetic stirrer 40mgr weight and it was continued to casting process.

III. Mathematics Method

In order to manufacturing hybrid MF membranes, the mathematic model of composites concept was used. The equation of composite is follow as below;

$$W_c = W_f + W_m \tag{1}$$

And the weight fraction (%wt) can be calculated based on the equation as below;

$$W_f = \frac{w_f}{w_c} = \frac{\rho_f}{\rho_c}.V_f \,, \qquad W_m = \frac{w_m}{w_c} = \frac{\rho_m}{\rho_c}.V_m \tag{2}$$

Therefore, the density of hybrid composite according to the weight fraction is calculated using equation:

$$\rho_c = \frac{1}{\left(W_f / \rho_f\right) + \left(W_m / \rho_m\right)} \tag{3}$$

Where; weight of reinforcement indicated is w_f, density of reinforcement is ρ_f. v_f is reinforcement volumes and V_f is volumes fraction.

In order to determine tension strength and modulus of elasticity of hybrid MF membranes the tensile test have been employed based the ASTM D 882. The strength and strain of membrane is calculated use the formula as below;

$$\sigma = \frac{P}{A} \tag{4}$$

$$\varepsilon = \frac{\Delta l}{l_o} \tag{5}$$

Then the modulus of elasticity of membranes is calculated using equation below;

$$E = \frac{\sigma}{\varepsilon} \tag{6}$$

where; P is loads (N), "A" is orthogonal space area (mm^2), "*lo*" is specimen initial length on tension area and length propagation of tension area of specimen is "*Δl*". Then tension strength is "*σ*"(N/mm^2), "*ε*" shows strain (mm/mm), and *E* is modulus of elasticity *(GPa).*

Afterwards; effect of absorption on the membranes, swelling was occurred. Swelling is explained as enlargement normal size of a material that caused by a numbers of water adsorption into materials. The swelling of hybrid micro membranes have been calculated using the equation:

$$Swelling\,(s) = \frac{W_{c\,wet} - W_{c\,dry}}{W_{c\,dry}}\, x100\% \quad (7)$$

Where; 'S" is identity of swelling membrane and W_{dry} is weight of specimen in dry condition; W_{wet} is specimen weight after the bath.

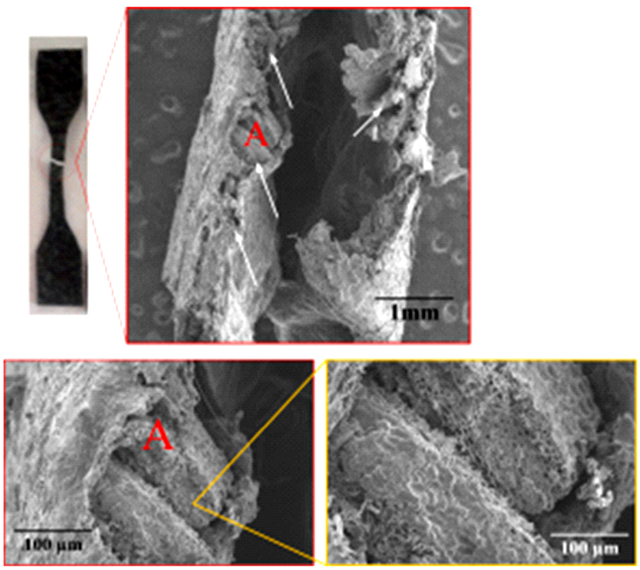

Fig. 4 SEM of hybrid MF membrane failure after tension test at low magnification.

Table 2
TENSION TEST VALUES OF MEMBRANES

Membrane classification	Cod e	Strength (N/mm2)	Strain	Modulus of elasticity (GPa)
MBH1	H1	1.42	0.48	0.003
MBH2	H2	1.15	0.55	0.002
MBH3	H3	0.61	0.78	0.0008
MBH4	H4	1.40	1.10	0.0012

IV. Result and Discussion

A. Tension failure of hybrid MF membranes

Fig. 4 show failure performance of hybrid MF membrane after the tension test. The failure of specimen was analyzed using SEM on low magnification. As shown on the Fig. bond intact was occurred between the particle and polymer. Letter "A" on Fig. show the moringa oleifera particle. Moringa has much porosity that indicates very significant in absorb the water pollutant. Also, between the particle and polymer do not occurred delaminate and pull-out. Agnihorti et.al [12] also explain that moringa oleifera has many porous like honey comb structure.

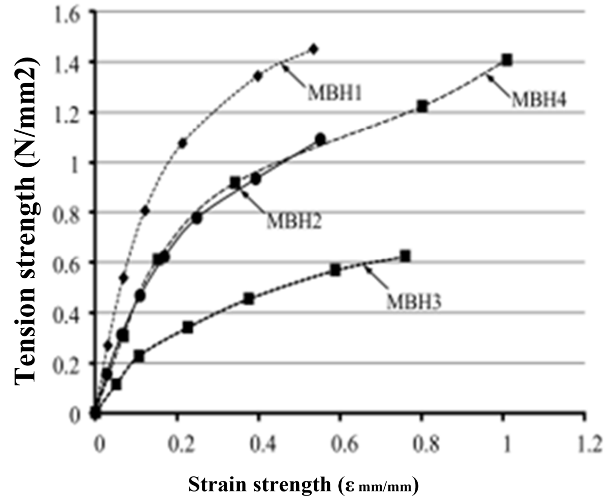

Fig. 5 Tension vs. Strain curve of Hybrid MF membranes

B. Mechanical properties of the hybrid MF membranes

Fig. 5 illustrates the correlation curve between the tensions versus strain each variation of specimen (MBH1, MBH2, MBH3 and MBH4). The values of every hybrid MF membranes are noted on Table.2. The highest value of tension strength is shown on MBH1, but it has low of strain value (ε =0.48). In addition, MBH4 with combination MO (23.1 wt %) /CA (23.1wt %) /PVA (70wt %) /SC (0wt %) has almost similar tension strength value with the MBH1. However, MBH4 shows longest strain value than other type of membrane (MBH1, MBH2 and MBH3). It is indicates that increase content of PVA increasing a ductility of membrane. Similar result was shown with [24]. Here, author explains that tensile strength shows resistance of membrane against the mechanical force and elongation that reflects the flexibility of membrane.

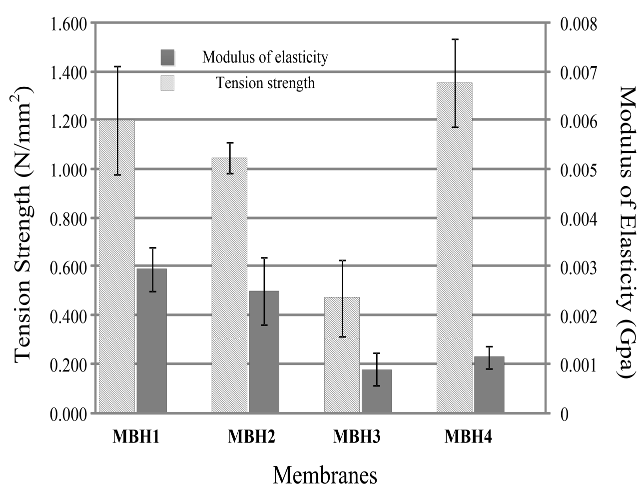

Fig. 6 Curve of tension strength and modulus of elasticity for hybrid MF membrane model.

Table 2 shows the tension, strain and the modulus of elasticity of hybrid MF membrane for each model. In this experiment, researcher have been agreed that the particulate reinforced composite is a less effective means of tension strength and the modulus of elasticity, respectively. It caused by the size of particle influences the bonding qualities. On the other hand, the PVA on dry condition has weak bonding to the particles, so membrane become brittle. The value of tension strength and modulus of elasticity of hybrid MF membrane are shown on Fig. 6. As shown on the Fig. the MBH1 and MBH2 hybrid MF membranes type has a high value the modulus of elasticity. In contrast, the MBH3 and MBH4 have low value the modulus of elasticity. And their tension strength has a highest value. This phenomenon is significantly influenced by contents value of ceresin and PVA on each specimen model.

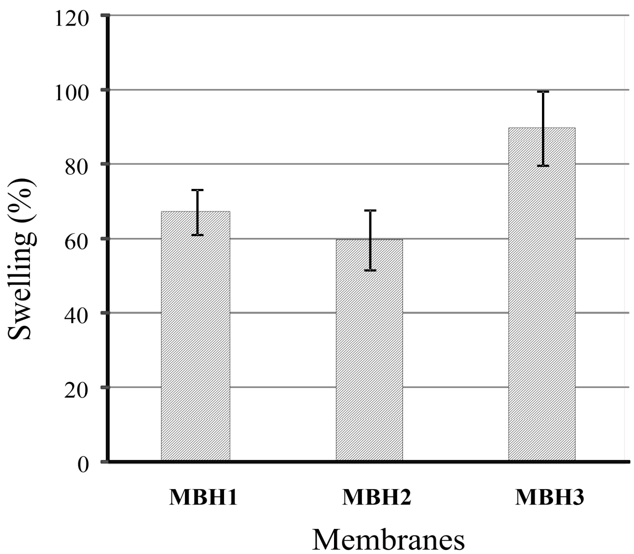

Fig. 7 Swelling characteristics of hybrid MF membrane

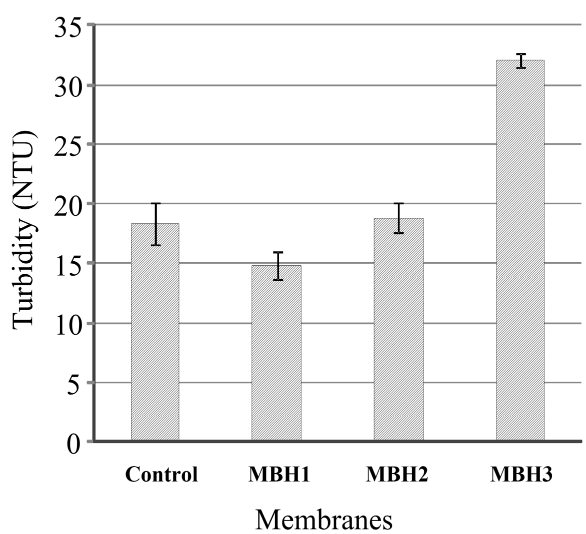

Fig. 8 Turbidity water of the hybrid MF membranes

C. Hybrid membranes on water purification.

Fig. 7 shows the swelling curve for each model membranes. Based on calculations using the equation (7) is obtained for each model of membrane swelling by 47.21%, 60% and 89.88%, respectively compared to membranes with dry conditions. The high value of the percentage of swelling of the membrane proves that the membrane is excellent in the water absorption and binding of pollutants contained therein. It is also influenced by the porosity of the hybrid MF membrane. In other words, these results indicate the existence of hydrophilic properties, so it is good for absorption membranes.

Fig. 8 shows the characteristics of the membrane towards the water turbidity. From the filtration process using a *dead-end filtration (DEF)* test, as long as 2 hours testing have generated some data as indicator of the cloudiness (turbidity) and data microbiological parameters (coli form and fecal coli form).

The turbidity value of MBH2 tends to increase 18.81 NTU. It is mean increased of 2% against the pre-treatment of raw water. The MBH3 has the highest turbidity values with an average value of 32.04 NTU, that mean increased of 42%. This is influenced by future soluble PVA [25], making the water becomes murky. Turbidity in the membrane MBH3 is also due to the low weight of the fraction of coconut shell activated carbon. In contrast to MBH1 with high carbon content capable of lowering the turbidity of raw water up to 19% of the control that the average value of 14.82 NTU.

V. Conclusion

Hybrid microfiltration membrane with particulate and biopolymers solutions were made for water purification. In this experiment composition between the particulate and biopolymer based the weight fraction ratio of 70%; 30% was carried out. This membrane was manufactured using the casting method accordance to ASTM D882 standard. The membrane thickness is 1.06~1.51 mm. Then tensile testing, swelling, and Dead-End Filtration (DEF) were conducted for test mechanical and physical membranes.

Following the result, water turbidity using the hybrid membrane microfiltration (MBH1) was shown lowest value compared with other hybrid membrane models. It caused by the MBH1 has highest number of activated carbon, and they will reduce the water color and smelt significantly. In contrast, decreasing number of activated carbon on hybrid MF membrane was increased the turbidity number of water. It was caused by biopolymer (PVA) has behavior easy melt in the water. Moringa oleifera have significant affect on decrease the number of chemical contained and microbiology in the water.

An experiment can be concluded that hybrid microfiltration membrane with reinforcement particulates (i.e. moringa and activated carbon coconut shell charcoal) and biopolymer (i.e. PVA and Ceresin) have potential for purification water. In this experiment all of material can be used for decrease the water pollutants contained in the same time.

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