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Fabrication by Electrospinning Technique and Characterization of Curcuma mangga Val Reinforced PVA Fibrous Membranes

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Abstract— Antimicrobial and anti-diabetic of Curcuma mangga Val properties have attracted research interest. Curcuma mangga Val extract (CME) and polyvinyl alcohol (PVA) were blended, and fabricated to the fibrous membranes by the electrospinning method. The effects of CME concentration on the fiber morphology and tensile properties of CME/PVA membranes are the goal of current study. Scanning electron microscopy (SEM) of the membranes revealed bead-free fibers with straightly/continuously orientation formed in all CME/PVA membranes. The average fiber diameter increased from 186 nm to 297 nm with the CME concentration from 0 to 3 wt.%, respectively. The addition of 1% CME resulted in relatively high tensile strength 24.96 ± 0.20 MPa, which is the highest among the CME/PVA membrane specimens. However, very high tensile modulus and low elongation showed in these results lead to reducing the functionality of the CME/PVA fibrous membrane due to the brittleness. The CME properties may contribute to those shortages.

Keywords— Curcuma mangga Val; PVA; electrospinning; fibrous membrane; tensile properties.

1. INTRODUCTION

Curcuma mangga Val is well known in Asia, especially Indonesia, and also in Africa and Australia. Curcuma mangga Val is a kind of plant that possesses the beneficial properties for medicine materials such as antimicrobial [1], anti-diabetic [2], and non-toxic. Therefore, numerous studies on Curcuma mangga Val have been carried out for medicine mostly in Indonesia. Those properties have appealed to biomedical research interest. It can be potential as an alternative wound dressing based material. In this case, Curcuma mangga Val extract (CME) would be useful as reinforcing material for the polymer as chitosan and aloe Vera to fabricate fibrous membranes by electrospinning technique.

Chitosan and aloe Vera are the natural materials having the properties of antimicrobial activity, anti-inflammation, and non-toxic [3, 4]. Those properties had also dramatically increased the interest to use them as based materials for wound healing, wound dressing, scaffold, and tissue engineering applications. The fibrous membranes made of chitosan and aloe Vera combined with poly (vinyl) alcohol (PVA) [5–7], polyethylene oxide (PEO) [8] and polycaprolactone [9] had been extensively investigated. However, no study of CME-based fibrous membranes fabricated by electrospinning method was found. Thus, it is a

great challenge to initially create the work to make the CME-based fibrous membrane by electrospinning. It would be a benefit in biomaterial science.

In the studies with electrospinning, PVA is frequently used due to its synthetic polymer nature, which is conductive and easily blends with chitosan as a natural polymer. The chitosan combined the PVA solution in various ratios of chitosan to PVA produced the chitosan/PVA nanofiber mats with differences in fiber structures corresponding to the ratio. A rise of chitosan content tended to form beaded fibers. A PVA/chitosan ratio of 50:50 showed severe bead formation [10]. The applied voltage in this study was also varied: i.e., 15 kV and 20 kV, and the fiber diameter changed irregularly in both conditions. The smallest fiber diameter (~ 148 nm) was achieved by the PVA/chitosan ratio of 70:30 at 20 kV. By a similar proportion and applied voltage of 20 kV, the chitosan/PVA exhibited the tensile strength of 5.26 ± 0.53 MPa, which was lower than that of neat PVA 7.4 ± 0.37 MPa [11].

On the other hand, the hybrid fibrous membranes of chitosan nano-emulsion blended with aloe Vera/PVA were prepared by the electrospinning method at an applied voltage of 15 kV. The membrane at 15% chitosan nano-emulsion concentration yielded the highest tensile strength of 6.18 ± 0.51 MPa with an average fiber diameter of 180 nm [6].

Except for the viscosity as a function of concentration, the electrical conductivity of the polymer solution has played a crucial role in the formation of fiber morphology as well [12–14]. Insufficient volume charge density in the polymer solution, the fiber would not be formed. As mentioned earlier, PVA is one of the conductive polymers. Based on our experience, the high molecular weight (Mw) of PVA was more challenging to blend with the reinforcing materials in comparison with the low Mw of PVA.

Due to no information of study on the Curcuma manga Val based electrospun fibrous membranes, in this preliminary study, CME concentrations were varied and incorporated into the PVA solution. To avoid the difficulty of polymer solution preparation, PVA used in this work was low Mw of PVA, which rarely used in other studies. Changes in the characteristics of CME/PVA fibrous membranes due to different CME concentrations were discussed. Besides, the results are compared to other fibrous membranes made of antibacterial natural materials reinforced PVA.

2. MATERIALS AND METHODS

Curcuma manga Val (GMV) (Fig. 1a) used in this research was a commercial Curcuma manga Val extract purchased at “Obat Herbal Alami Jogja” shop, Yogyakarta, Indonesia. PVA Gohsenol (PVOH/PVA, Mw: ~ 22,000 g/mol) purchased at CV. Multi Kimia, Yogyakarta, Indonesia, was used.

The manufacturing of the fibrous membranes was begun by preparing a PVA solution with 10% concentration: i.e., by dissolving 10 g PVA powder into 100 g H₂O by continuously magnetic stirred at $80 \pm 2^\circ\text{C}$ for one hour. The CME powder with a particle size of 400 mesh (Fig. 1b) was subsequently dissolved into the PVA solution with varying CME concentrations of 0%, 1%, 2%, and 3%, and magnetically stirred continuously at 200 rpm and 50°C for one hour. After that, the CME/PVA solution was left for four days (Fig. 2), which was used for the spinning solutions or feed of the electrospinning process. The viscosity and electrical conductivity of the spinning solutions at each CME concentration were measured. The CME/PVA fibrous membranes were fabricated at the optimized condition of applied voltage of 18 kV, a distance from a tip to the collector (TCD) of 15 cm, the diameter of spinneret 0.6 mm and flow rate of 0.003 ml/min. The fibrous membrane specimens were designated as CME-0/PVA, CME-1/PVA, CME-2/PVA, and CME-3/PVA. In this case, the optimization of applied voltage and TCD was carried out by fabricating thin fibrous membranes (Table 1) at various applied voltage and TCD. The fiber structure formed in the membranes was then observed under an optical microscope and optimized. The optimum condition was selected based on the uniform fiber orientation with free fibers-defects or free from beads formation. This is shown in an optical micrograph resulted at applied voltage and TCD of 18 kV and 15 cm, respectively (Table 1).

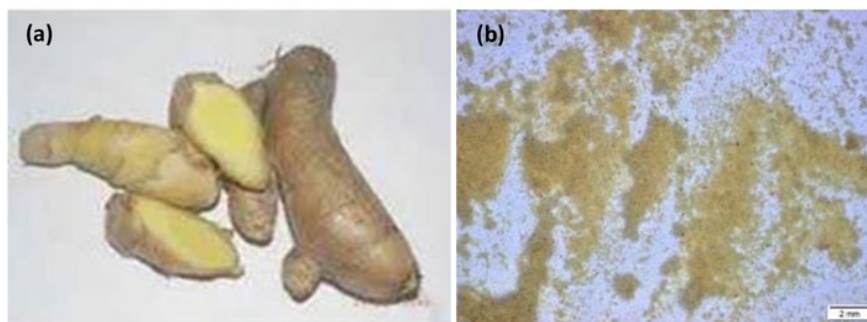


Fig. 1 The photographs of Curcuma manga Val (a), and curcuma manga Val extract (CME). (b).

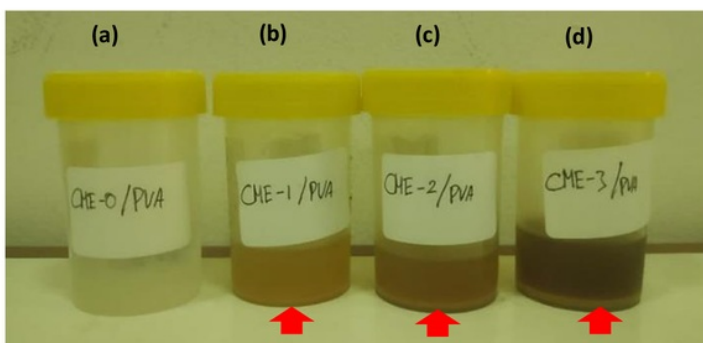
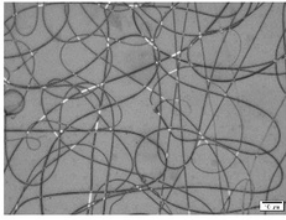
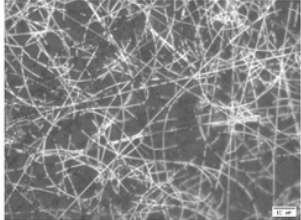
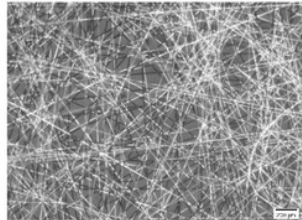


Fig. 2 The spinning solutions of PVA (a), and CME_PVA with different CME concentrations, 1% (b), 2% (c) and 3% (d) showing the presence of precipitation (see, red arrows).

TABLE I
THE FIBROUS MEMBRANES FORMED AT SOME DIFFERENCES IN APPLIED VOLTAGE AND TCD

Applied voltage and TCD		
15 kV and 13 cm	15 kV and 15 cm	18 kV and 15 cm
		

The characterization of fiber morphology built-in CME/PVA fibrous membranes was performed by scanning electron microscopy (SEM, Hitachi SU-3500). The measurement of fiber diameter was conducted using the ImageJ digital image analysis on at least one hundred points.

The tensile test was conducted on all fibrous membrane specimens according to ASTM 882 using a universal testing machine (UTM, Zwick Z0.5 Germany) at a crosshead speed of 10 mm/min and a specimen gauge length of 20 mm. In each case, eight specimens were prepared and tested.

3. RESULT AND DISCUSSION

3.1 Morphology

SEM images of all fibrous membrane specimens (Fig.3) exhibit a relatively similar fiber structure: i.e., bead-free and straightly oriented. However, the average fiber size appeared differently. A rise in CME concentration increases the fiber size, as exhibited in the distribution of fiber size (Fig.4). The fiber size in a neat PVA fibrous membrane (CME-0/PVA) is mostly present in the range of 100 – 200 nm. The fibrous membranes of CME-1/PVA and CME-2/PVA show nearly similar fiber size distribution in the between 200 – 300 nm, but a portion of fiber size at the position between 100 – 200 nm in the CME-1/PVA membrane is higher than that in the CME-2/PVA membrane. The largest fiber size is mostly in the range between 200 – 300 nm and 300 – 400 nm demonstrated in a CME-3/PVA membrane. Enhancement of a mean fiber size by increasing the CME concentration is associated with an increase of the viscosity of the spinning solution (Table 2). It also shows that an increase in CME concentration decreases the electrical conductivity. It might be owing to the un-ionic nature of the CME. It shows that an increase in CME concentration decreases the electrical conductivity. It might be due to the un-ionic nature of the CME. The trend of those results is consistent with that reported by Shahreen et al. [14]. In this study, however, the increasing viscosity and reducing the electrical conductivity affected by an increase of CME concentration did not lead to the formation of beads. The existence of beads in the fiber would inhibit the interaction between the fibers, which

usually lead to reducing the tensile strength value of the membrane [15].

In the electrospinning process, the viscosity and electrical conductivity are the vital parameter processes affecting the fiber formation. The polymer solution will be suitable for electrospinning when the polymer solution viscosity is present between 100 cP – 2000 cP [12]. Based on our experience, the solution having low viscosity leads to drop occurrence and then usually produce beaded-fibers. Too high viscosity, the polymer solution will be difficult to pump through the syringe needle, and the solution may dry before starting the electrospinning process. Usually, the fiber size increased.

Thus, the excellent fiber structure resulted in the current work might be caused by the viscosity of all CME_PVA solutions that are in the range between 100 cP and 2000 cP. However, the formation of bead-free and continuous fibers in polyethylene oxide (PEO) fibrous membrane occurred at the viscosity of 1250 cP. The bead-free and continuous fibers built in the hybrid membrane of CSNe/AV_PVA with the average fiber size of 242 nm formed by the polymer solution viscosity of 379.9 cP [5]. Those in the PdCl₂/polyvinylpyrrolidone (PVP)/titanium (IV) isopropoxide (TTIP) fibrous membranes revealed at the viscosity of 65.8 cP with 256 nm fiber size [14]. Besides, a study on the fibrous membranes made of mixed (PVA and chitosan) solutions has verified the changes in both the average fiber diameter and fiber structure. Those are affected by the volume ratio of PVA to chitosan, and also the applied voltage. In this case, the viscosity of mixed PVA and chitosan solutions are a function of the volume ratio, although the effect did not change linearly [10]. A correlation between the polymer solution viscosity and concentration is strictly dependent on the nature of the polymer and the intermolecular interactions within the polymer solution [17]. In principle, to produce the fibers by electrospinning technique is that the polymer solution should have sufficient molecular weight, viscosity, and also electrical conductivity [12, 13].

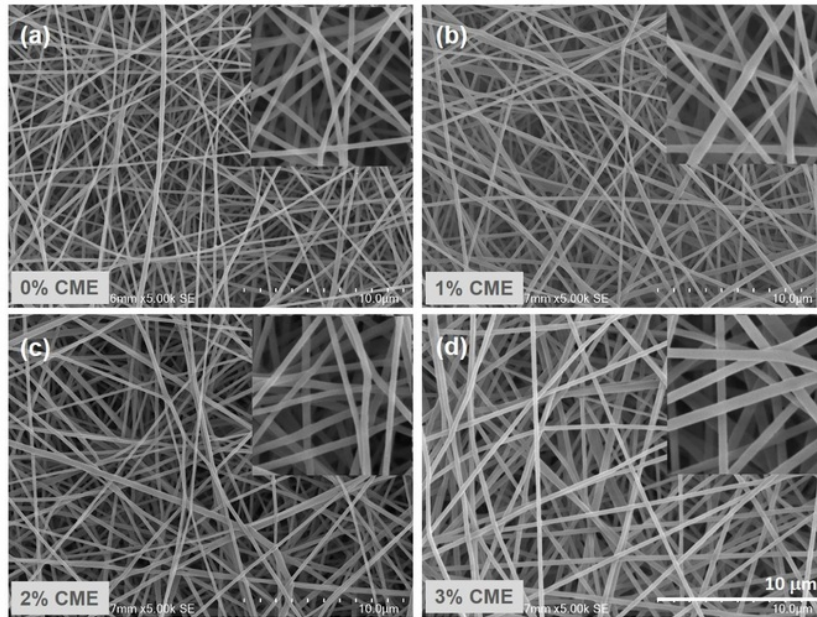


Fig. 3 SEM images of CME/PVA nanofibrous membranes with differences in CME concentrations. (a) 0% CME, (b) 1% CME, (c) 2% CME and (d) 3% CME.

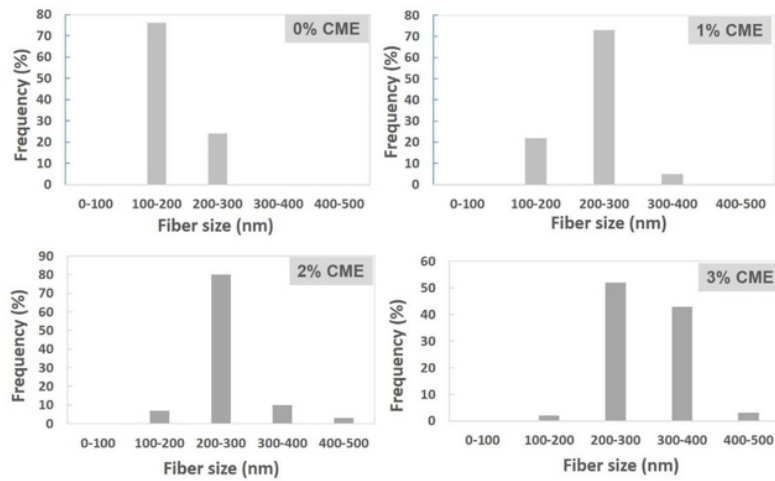


Fig. 4 Fiber size distribution related the SEM images in Fig.3a, 3b, 3c and 3d, respectively.

TABLE III
VISCOSITY, ELECTRICAL CONDUCTIVITY AND AVERAGE FIBER SIZE OF THE CME/PVA FIBROUS MEMBRANES

No	Specimen	Viscosity (cP)	Electrical conductivity ($\mu\text{s}/\text{cm}$)	Average fiber size (nm)
1	CME-0/PVA	436	451	186.21
2	CME-1/PVA	452	423	227.07
3	CME-2/PVA	466	393	251.04
4	CME-3/PVA	482	340	296.67

3.2 Tensile Properties

On the other hand, the fiber morphology resulted in all fibrous membranes may correspond to their tensile properties (Fig. 4 and Fig.5). Insertion of 1% CME in the PVA solution improves the tensile strength of CME-1/PVA fibrous membrane, but further increases to 2% and 3% CME reduced the tensile strength of the fibrous membranes. The tensile strength obtained from this result is quite high; it is higher compared to other findings [5, 11, 15, 17]. However, the tensile modulus of all fibrous membranes is also remarkably high. They are too high; thus, the membranes tend to be brittle, although the tensile strengths are considerably high. According to the result of the tensile-stress versus strain at break (Fig. 5), it indicates that maximum elongation reached by the neat PVA fibrous membrane. The addition of CME decreased elongation. The higher the CME concentration, the lower the elongation.

In comparison with a study on the fibrous membranes made of aloe vera extract (AVE) reinforced PVA [5], at the similar concentration of 2% AVE, the tensile properties showed 5.74 MPa for tensile strength, 33.99 MPa for tensile modulus, and 135% for elongation at break with the average fiber size of around 342 nm. The fiber structures resulted from the current work are much better, and the average fiber

size is also smaller compared to those resulting from the AVE/PVA fibrous membrane.

Based on those results, the predominant factor influencing the tensile properties of the CME/PVA membrane, especially the improvement of the tensile modulus and reducing the elongation, is of the properties of CME particles rather than the fiber size and structure. The commercial CME particle size might be too coarse and impure, leading to form the precipitation (Fig. 2). In this case, CME could not be dissolved in the PVA solution completely. Thus, the particle size should be down to a nanometer to disperse the CME particles in the PVA solution uniformly. Another propose for an alternative way is to prepare CME by making the Curcuma mangga Val filtrate and then blend with the PVA solution. Therefore further research on the electrospun fibrous of CME/PVA is necessary because of the valuable properties of Curcuma mangga Val. All CME/PVA fibrous membranes produced from this preliminary study using CME, however, could not be recommended as an alternative wound dressing material. The stiffness and brittleness were quite high, although the current work achieved very high tensile strength and excellent fiber morphology built in the membranes.

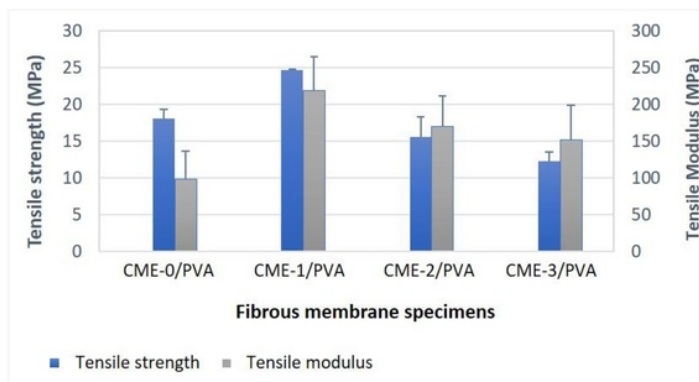


Fig. 5 Tensile strength and modulus of CME reinforced PVA fibrous membranes with various CME concentrations.

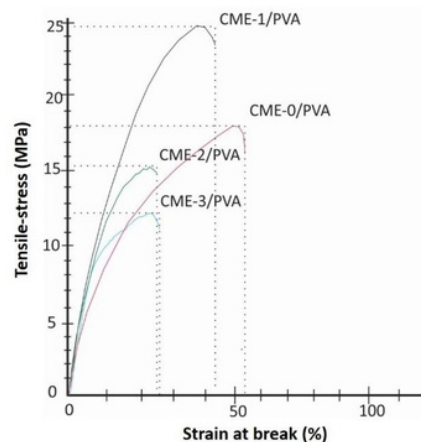


Fig. 6 Tensile-stress versus strain at break.

4. CONCLUSIONS

These experimental results have provided new information on the electrospun fibrous membrane made of Curcuma mangga Val reinforced PVA. The fibrous membranes have successfully fabricated and yielded comparatively small fiber size with bead-free continuous fibers. Those contributed to the improvement of the tensile strength CME/PVA fibrous membranes, especially on the membrane, by adding 1% CME (24.96 ± 0.20 MPa). However, all the produced CME/PVA membranes are being less functional material due to their very high tensile modulus and low strain. These conditions made the membrane tend to be brittle. The properties of CME might be the cause. The shortcoming of these results would be corrected by developing the preparation technique of CME in the next research.

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