Synthesis and Characterization of Cellulose–Curcumin Nanofiber as a Biomaterial Mask

Novrynda Eko Satriawan1*, Muhamad Nasir2, Fasih Bintang Ilhami3

1Department of Chemistry, Institut Sains dan Teknologi Annuqayah, East Java, Indonesia
2Research Center for Clean Technology, National Innovation and Research Agency, Bandung, West Java, Indonesia
3Institute of Atomic and Molecular Science, Academia Sinica, Taipei, Taiwan
*Corresponding author: nekosatriawan@istannuqayah.ac.id

Received: 11 September 2021; Accepted: 30 June 2022; Published: 15 July 2022

Abstract

This study was conducted to characterize cellulose acetate-curcumin composite nanofibers synthesized using the electrospinning method, as a biomaterial mask with antibacterial and antiviral properties. Cellulose acetate-curcumin composite nanofibers were successfully synthesized at a flow rate of 1 mL/hour, a needle-collector distance of 17 cm, and a voltage of 22 kV. The resulted nanofibers have an average diameter in the range of 378.89 nm - 461.76 nm. Based on the results of the FTIR spectrum, no significant shift was found. The results of the contact angle test showed that the average contact angle value increased with the addition of curcumin concentration. The mechanical test results showed that variations in the concentration of curcumin were able to increase the tensile strength and strain values.

Keywords: nanofiber; cellulose acetate; curcumin; mask; electrospinning

Introduction

Covid-19 has attacked in every country in this world. To prevent the spread of COVID-19, various methods have already been used, including regional closings and isolation, imposing curfews, tightening activities that involve large groups of people, social distancing programs, and the required wearing of masks for everyone who engages in outdoor or indoor activities. The spread of COVID-19 occurs when there is direct interaction with a suspected patient or asymptomatic person. Transfer can occur through airborne droplets produced from saliva when sneezing, coughing, or talking (di Gennaro et al. 2020; Shereen et al. 2020).

Masks are one of the policies that achieve a balance between enabling individuals to move while also ensuring that economic activity does not stop. By restricting the flow of droplets from suspected or asymptomatic persons in the air and reducing the probability of a person’s hands directly touching the mouth and nose, masks significantly reduce the transmission rate of COVID-19 (Cheng et al. 2020).

Masks can act as a first line of defense against virus-infected droplets. During a pandemic, masks are one of the most crucial parts of medical equipment. The creation of mask items manufactured from natural substances that also have antibacterial and antiviral properties could be an alternate strategy for increasing effectiveness in treating the COVID-19 virus’s spread pattern. The use of biodegradable natural materials with antibacterial and antiviral properties...
will add value to the masks production and may improve their performance.

Cellulose acetate is an ester of cellulose, a natural polymer that is plentiful in nature, sustainable, and can be degraded naturally (Huang & Dean 2020). Cellulose acetate has a high attractiveness due to its biodegradable nature, so that it is environmentally friendly, biocompatible, having good affinity with other substances, and having good modulus, flexural strength, and tensile strength.

Antimicrobial membranes, bladder matrix materials, biosensors, and drug delivery systems are all examples of cellulose acetate's hydrophobic properties. It’s also easy to shape, doesn’t wrinkle easily, and has a high stability. Therefore, it's widely used in pharmaceutical and biomedical fields. (Yu et al. 2012).

The major ingredient of turmeric (Curcuma Longa L.), a natural chemical widely used in traditional medicine, is curcumin. Curcumin possesses antioxidant, analgesic, anti-inflammatory, antiseptic, anticarcinogenic, antibacterial, and antiviral effects, among others (Chh-Hung & Ann-Lii 2007; Mathew & Hsu 2018; Mutlu et al. 2018; Li et al. 2019). Biomaterials with antibacterial and antiviral properties could be cellulose acetate–curcumin composites. The addition of curcumin to the cellulose acetate polymer can boost the material’s antioxidant properties (Xie et al. 2020).

Fibers with a diameter of less than 1 micron are known as nanofibers. Interesting qualities emerge when the diameter of the fiber decreases from micrometer to nanometer, such as a larger surface-to-volume ratio (10^3 times larger than a micrometer-sized fiber), increased flexibility and mechanical strength, and increased porosity (Majumder, Sharif & Hoque 2020). Electrospinning is a technique for producing nanofiber sheets that uses high-voltage electrical energy.

Electrospinning nanofibers have several advantages, including a large surface area, a pore structure with large porosity, a wide range of diameters that can be modified, ease of usage, and versatility (Celik & Oksuz 2015). Because bacteria have diameters in the 1000 nm range and viruses have sizes in the 100 nm range, using nanofibers as filters on masks could be an excellent option for enhancing mask effectiveness. This study should yield preliminary results in the development of electrospinning methods for producing nanofibers as the basis of antiviral and antibacterial masks.

**Research Methodology**

**Materials and Tools**

The materials used in this study were cellulose acetate (Mw 30.000 Sigma Aldrich), Curcumin (Merck), Acetone p.a. (Merck), N,N-Dimethyl Acetamide (DMAc) (Merck). Beside, the instruments used are a set of electrospinning equipment, SEM JSM-IT300, FTIR Prestige 21 Shimadzu, a set of tensile test equipment Universal Testing Machine (UTM) Shimadzu AGS-X series 10 kN, measuring flask, beaker, measuring pipette, measuring cup, Magnetic Barr, Magnetic Stirrer, Electric Balance, Box Spuit + Needle Terumo.

**Work procedures**

Cellulose acetate polymer solution was prepared with a concentration of 17% using acetone/DMAc solvent with a ratio of 2:1. A cellulose acetate–curcumin composite solution was prepared by mixing curcumin with a concentration variation of 2.5%, 5%, 7.5%, and 10% w/w in the 17% cellulose acetate polymer solution. The solution was then stirred for 4 hours until homogeneous. The electrospinning process was carried out with variations in flow rate, needle distance to the collector, and voltage.

**Data analysis**

Observational data consisted of several types, namely surface morphology observation data using FE-SEM, mechanical test data, FTIR test results, and data on the contact angle of the liquid against the resulted nanofiber.
Results and Discussion

Based on the research that has been done, data obtained from the observation of the morphology of cellulose acetate nanofibers with a concentration of 17% (Satriawan 2017), which was carried out using a Scanning Electron Microscope (SEM). The test was carried out on samples that were varied based on the voltage value in the electrospinning process, such as at a distance collector of 17 cm, flow rate of 1 mL/hour, and voltages of 21 kV, 22 kV, and 23 kV. The best fiber morphology in the electrospinning process with a collector distance of 17 cm, a flow path of 1 mL/hour, and a voltage of 22 kV the best fiber morphology in the electrospinning process is similar to the optimum conditions obtained by the study (Satriawan 2017).

A scanning electron microscope (SEM) was used to examine the morphology of cellulose acetate-curcumin composite nanofibers. The testing was carried out on samples by using an electrospinning technique with a 17-cm collector distance, a 1 mL/hour flow rate, and a voltage of 22 kV. Figure 1 shows the findings of observations made by using a scanning electron microscope (SEM).

Figure 1. The results of SEM observations of cellulose acetate nanofibers at a collector distance of 17 cm, a flow rate of 1 mL/hour, and a voltage of 21 kV (a), 22 kV (b), and 23 kV (c)

Figure 2. The results of SEM observations on cellulose acetate-curcumin composite nanofibers at a collector distance of 17 cm, a flow rate of 1 mL/hour and a voltage of 22 kV with a curcumin concentration variation of 2.5% w/w (a); 5% w/w (b); 7.5% w/w (c); and 10% w/w (d)

Based on the results of observation, the surface morphology in more detail and the diameter of the resulted fiber can be seen in the following Figure 2.

Follow-up observations were made to determine the morphology of the fiber more clearly. Observations were made at 20,000 times magnification to see the surface morphology and pores that might form on the bars of the resulted fiber. The surface of the resulted fiber can represent the homogeneity of the mixing and dissolving process of the composite between cellulose acetate and curcumin polymers at various concentrations.
of the composite solution, as shown in the following Figure 3.

The results of observations using SEM obtained data showed that the nanofiber produced was very good, which was marked without any beads or bits. With a layered matrix structure, the nanofibers formed are elongated and continuous. The homogeneity of the solution resulted from the mixing and dissolving process of the composite between cellulose acetate polymer and curcumin as a charge can be represented by the resulted nanofiber surface, which indicates that there has been good dissolution so that the curcumin blends with the cellulose acetate polymer in each resulted nanofiber.

Based on the diameter distribution of the resulted nanofibers, cellulose acetate nanofibers have an average diameter of 378.89 nm, cellulose acetate-curcumin nanofibers 2.5 % w/w have an average diameter of 457.25 nm, cellulose acetate-curcumin nanofibers 5.5 % w/w have an average diameter of 408.39 nm, cellulose acetate-curcumin nanofibers 7.5 %, and cellulose acetate-curcumin nanofibers 10 % w/w have an average diameter of 461.76 nm.

Cellulose acetate nanofibers have non-uniform diameters. The diameter distribution of the resulted nanofibers is mostly in the range of 300–399 nm. Cellulose acetate nanofibers 2.5% w/w have diameter distributions in the range of 300–499 nm. Cellulose acetate nanofibers-curcumin 2.5% w/w have diameter distributions in the range of 300–499 nm. In the range of 200–499 nm, cellulose acetate-curcumin nanofibers 7.5% w/w have the most diameter distribution, followed by cellulose acetate-curcumin nanofibers 10 % w/w.

The complete diameter distribution of the resulted nanofibers is presented in the diameter distribution graph in the following Figure 4.

The cellulose acetate fiber was observed by using the Fourier Transform Infrared (FTIR) Prestige Shimadzu 2.1. It’s to determine the specific functional groups, shifts, and changes in the intensity of the absorption band of cellulose acetate fiber before and after the addition of curcumin. As shown in Figure 5, the FTIR results of the cellulose acetate fiber samples were compared to the FTIR results of the cellulose acetate-curcumin composite nanofiber samples.

Figure 3. The results of SEM nanofiber observations at 20,000 times magnification, with a collector distance of 17 cm, a flow rate of 1 mL/hour, and a voltage of 22 kV (A) CA; (B) CA-Cur 2.5% w/w; (C) CA-Cur 5% w/w; (D) CA-Cur 7.5% w/w; and (D) CA-Cur 10% w/w
Figure 4. FTIR results from cellulose acetate nanofiber samples and cellulose acetate-curcumin composites at various concentrations

Figure 5. FTIR results from cellulose acetate nanofiber samples and cellulose acetate-curcumin composites at various concentrations

Figure 6. Tensile test results of cellulose acetate nanofiber samples and cellulose acetate-curcumin composites at various concentrations
The cellulose acetate molecular formula is \((\text{C}_6\text{H}_7\text{O}_2(\text{OCOCH}_3))^n\), and the functional groups contained in the cellulose acetate molecule include OH groups, ester groups, and alkyl groups. Because curcumin has a functional group similar to cellulose acetate, there it seems be no difference in the FTIR analysis results. Based on the results of the comparison, there was no significant shift in the FTIR spectra produced by the cellulose acetate polymer and the cellulose acetate curcumin composite at various concentrations. Cellulose acetate and curcumin have similar functional groups, and there is no specific distinguishing group between them.

The contact angle value is required to determine the properties of the material to be used as an antibacterial and antiviral mask material. The hydrophobic nature of the material will give a good value, indicating that water particles will not easily penetrate the material. Because the COVID-19 virus spreads through water droplets, the hydrophobic nature of the material will be able to resist the entry of virus-containing droplets into the material's interior.

Based on the results of the contact angle test at various concentrations of cellulose acetate-curcumin composite nanofibers, the average contact angle for cellulose acetate was 110°, cellulose acetate-curcumin 2.5% was 109°, cellulose acetate-curcumin 5% was 118°, cellulose acetate-curcumin 7.5% was 117°, and cellulose acetate-curcumin 10% was 122°.

Contact angle testing was carried out by using J Image software with the addition of a plugin contact angle. According to (Lamour et al. 2010), Droplets of a liquid on the surface of a solid can show the value of the adhesion force between the liquid and the solid surface, which results in the spread of the liquid (wet) and the cohesive force between the molecules of the liquid, which negates the dispersion of the liquid.

When a liquid is polar, it forms a contact angle when it is dropped on a non-polar solid and vice versa. If a polar liquid is dropped on a polar plate, a contact angle will not be formed. Contact angle can occur because of the adhesion force between the solid and the liquid and the cohesion force between the liquid molecules. The smaller the contact angle obtained, the greater the adhesion force. The greater the contact angle obtained, the greater the cohesive force (Fritzsche & Peuker 2015).

Based on the results of the contact angles, it is known that the distilled water and the surface of the cellulose acetate and cellulose acetate-curcumin nanofibers produced a large cohesive force. The greater
the cohesion force, it indicates that there is a polarity difference between them.

Analysis of mechanical properties was carried out to determine the values of tensile strength, stretch (elongation), and modulus of elasticity of cellulose acetate nanofiber membranes and cellulose acetate-curcumin composite nanofibers. The analysis results of the tensile strength and strain values using the results from mechanical testing, were showed in the graph of the analysis of the mechanical properties like in the Figure 6.

The test results showed that variations in the concentration of curcumin influenced the tensile strength and strain values of the cellulose acetate nanofibers and the resulted curcumin-cellulose acetate composites.

As for the highest tensile strength, it was at the concentration of curcumin addition of 7.5% w/w, while the highest strain value was at the concentration of addition of curcumin of 5% w/w.

Based on the test results in Figure 7 that the addition of curcumin to cellulose acetate affects the tensile strength of the resulted nanofiber. The highest tensile strength value was at the concentration of curcumin addition of 7.5% w/w with a tensile strength value of 0.71 MPa, while the lowest tensile strength value was in cellulose acetate nanofiber with a tensile strength value of 0.52 MPa.

As for the value of the modulus of elasticity, there was a significant change and increased with increasing concentration of curcumin at a value of 5% w/w of 0.12 MPa and 7.5% w/w of 0.16 MPa, with the highest value of with a concentration of 7.5% w/w addition of curcumin. The tensile strength and modulus of elasticity of the nanofiber can be seen in the following Figure 7.

Conclusions

Nanofiber composite cellulose acetate-curcumin was successfully made by the electrospinning method with a good surface morphology characteristic at a collector distance of 17 cm, a flow rate of 1 mL/hour and a voltage of 22 kV. The addition of curcumin to cellulose acetate influences the contact angle value of the resulted nanofiber. The highest contact angle value was in the addition of curcumin at a concentration of 10% w/w with an average value of 122.42°. The value of the concentration of curcumin added to cellulose acetate affects the tensile strength of the resulted nanofiber. The highest tensile strength value was at the addition of curcumin at a concentration of 7.5% w/w with a tensile strength value of 0.71 MPa. The lowest tensile strength was in the cellulose acetate nanofiber with a tensile strength value of 0.52 MPa.

Acknowledgment

This research was funded by the DIPA of the Directorate General of Higher Education, Ministry of Education, Culture 2021 and was supported by the research facility of the Clean Technology Research Workshop of the National Innovation and Research Agency in Cisitu Sangkuriang Bandung, West Java.

Data analysis was supported by research facilities, and scientific and technical support from the Cibinong Advanced Characterization - Integrated Bioproduct Laboratory at the National Research and Innovation Agency and one of the SEM images was taken at the SEM Laboratory of FMIPA ITB.

References

Celik, G. & Oksuz, A.U., 2015, "Controlled release of ibuprofen from electrospun biocompatible nanofibers with in situ QCM measurements," Journal of Macromolecular Science, Part A: Pure and Applied Chemistry, 52(1), 76–83.

Cheng, V.C.C., Wong, S.C., Chuang, V.W.M., So, S.Y.C., Chen, J.H.K., Sridhar, S., To, K.K.W., Chan, J.F.W., Hung, I.F.N., Ho, P.L. & Yuen, K.Y., 2020, “The role of community-wide wearing of face mask for control of coronavirus disease 2019 (COVID-19) epidemic due to SARS-CoV-2,” Journal of Infection, 81(1), 107–114.

Chh-Hung, H. & Ann-Lii, C., 2007, “Clinical studies with curcumin . PubMed Commons," The Molecular Targets and Therapeutic Uses of Curcumin in Health and Disease, 12, 471–480.
Fritzsche, J. & Peuker, U.A., 2015, *Wetting and adhesive forces on rough surfaces - An experimental and theoretical study*, Procedia Engineering, vol. 102, 45–53, Elsevier Ltd.

Gennaro, F. di, Pizzol, D., Marotta, C., Antunes, M., Racalbuto, V., Veronese, N. & Smith, L., 2020, “Coronavirus diseases (COVID-19) current status and future perspectives: A narrative review,” International Journal of Environmental Research and Public Health, 17(8).

Huang, H. & Dean, D., 2020, “3-D printed porous cellulose acetate tissue scaffolds for additive manufacturing,” Additive Manufacturing, 31, 100927.

Lamour, G., Hamraoui, A., Buvallo, A., Xing, Y., Keuleyan, S., Prakash, V., Eftekhar-Bafrooei, A. & Borguet, E., 2010, “Contact angle measurements using a simplified experimental setup,” Journal of Chemical Education, 87(12), 1403–1407.

Li, H., Zhong, C., Wang, Q., Chen, W. & Yuan, Y., 2019, “Curcumin is an APE1 redox inhibitor and exhibits an antiviral activity against KSHV replication and pathogenesis,” Antiviral Research, 167(December 2018), 98–103.

Majumder, S., Sharif, A. & Hoque, M.E., 2020, “Electrospun Cellulose Acetate Nanofiber: Characterization and Applications,” Advanced Processing, Properties, and Applications of Starch and Other Bio-Based Polymers, pp. 139–155, Elsevier.

Mathew, D. & Hsu, W.L., 2018, “Antiviral potential of curcumin,” Journal of Functional Foods, 40(December 2017), 692–699.

Mutlu, G., Calamak, S., Ulubayram, K. & Guven, E., 2018, “Curcumin-loaded electrospun PHBV nanofibers as potential wound-dressing material,” Journal of Drug Delivery Science and Technology, 43, 185–193.