



## Calculate The Conductivity of Some Composites of Cellulose Bacteria Mixed with Polypyrrol

Syukri Yunus<sup>1</sup>, Hairul Abrar<sup>2</sup>, Auliya Akbar<sup>1</sup>

<sup>1</sup> Department of Electrical Engineering, Faculty of Engineering, Andalas University, Limau Manis Padang

<sup>2</sup> Department of Mechanical Engineering, Faculty of Engineering, Andalas University, Limau Manis Padang

### ARTICLE INFORMATION

Received: July 01, 2021

Revised: November 15, 2021

Available online: November 30, 2021

### KEYWORDS

Composites, Bacteria Cellulose, TEMPO, Gambier, Polypyrrole, Conductivity.

### CORRESPONDENCE

Phone: +62 8363188688

E-mail: [syukriyunus@eng.unand.ac.id](mailto:syukriyunus@eng.unand.ac.id)

### A B S T R A C T

The manufacture of composites that have good electrical properties is to use a conductive polymer matrix. A conductive polymer is a polymer compound that has a stable bond that allows the polymer to act as a good conductor of electricity. This study aims to determine the highest conductivity value of composite materials that have been coated with polypyrrole, namely bacterial cellulose with polypyrrole (bio composite 1), tempo bacteria cellulose with polypyrrole (bio composite 2), and Gambier bacteria cellulose with polypyrrole (bio composite 3). In this study, there were four samples consisting of nata de coco (cellulosic bacteria), 2, 2, 6, 6-tetramethylpiperidine 1-oxyl (TEMPO), Gambier extract, and polypyrrole. Measurement of resistance value using the two point probe method. The results of this study obtained that the resistance and conductivity values of bio composite 1, bio composite 2, and bio composite 3 were 29.742 kΩ and 1.178×10<sup>-3</sup> S/cm, 20.338 kΩ and 1.692×10<sup>-3</sup> S/cm, 34,572 kΩ and 0.9807×10<sup>-3</sup> S/cm. The measurement results show that the highest conductivity value is bio composite 2.

### INTRODUCTION

Composite material is a material made from mixing two or more materials that have the mechanical properties of the constituent materials. Cellulose is one of the materials that have been used in the composite manufacturing process because of its flexibility [1].

Several bacteria are known to produce cellulose, but only *Acetobacter xylinum* bacteria can produce cellulose in sufficient quantities to meet market needs [3]. Bacterial cellulose has the same chemical structure as cellulose derived from plants, but bacterial cellulose has advantages such as high purity, high degree of crystallinity, high tensile strength, elasticity, and biodegradability [4].

The cellulose used in this research is nata de coco. Nata de coco or also known as bacterial cellulose is an alternative source for the supply of cellulose [5]. Nata de coco is produced by fermenting coconut water with the help of *Acetobacter xylinum*. Nata de coco is transparent, has a chewy texture resembling a gel and floats on the surface of the liquid. Nata de coco has a protohybril structure. Bacterial cellulose protofibrils

bind together like ribbons to form micro fibrils with a diameter of 20-50 nm. the tape has a thickness of 3-4 nm, a width of ~80 nm and a length of 1 – 9 m. Excellent bands of bacterial cellulose form a dense network structure. The structure of these protofibrils is what makes nata de coco properties have a high tensile strength after the water is separated [6, 7].

Bacterial cellulose has the advantages of high purity, high degree of crystallinity, has a density between 300 and 900 kg/m<sup>3</sup>, high tensile strength, and elasticity [8].

The manufacture of composites with good electrical properties generally uses a conductive polymer matrix. A conductive polymer is a polymer compound that has a stable (conjugated) bond. This allows the polymer to act as a good conductor of electricity. Among the conductive polymers, Polyethylene terephthalate (PET), Polypyrrole (Polipyrrole), and Polyaniline (PANi) are the types of conductive polymers that are widely used in research because they have the advantages of being easy to synthesize, chemical stability in good air and high electrical conductivity [9].

Polypyrrole is a conductive polymer that has several advantages such as being environmentally friendly, easy to

synthesize, has good redox properties, is easily electro synthesized in aqueous solution and has a relatively high conductivity compared to other conductive polymers [10, 18].

TEMPO which is used to oxidize can also be an efficient approach to weaken hydrogen bonds in bacterial cellulose and separate bacterial cellulose into thinner nanofibers resulting in better dispersion performance [11].

Various methods have been used in the manufacture of Nano cellulose, such as mechanical (high pressure), chemical (acid hydrolysis and TEMPO oxidation) and enzymatic methods [12]. The method with acid hydrolysis will remove the amorphous part of cellulose and Nano cellulose obtained has the properties of crystallinity, large surface area and high mechanical strength [13].

Gambier contains natural polyphenolic compounds in the form of catechins (7-33%) which have error groups, such as galocatechins and gall catechins. The chemical content of gambier other than catechins is tannic catechu acid (20-55%), pyro catechol (20-30%), fluorescent Gambier (1-3%), red kateku (3-5%), quercetin (2-4%), certain oils (1-2%), waxes (1-2%), and small amounts of alkaloids [14].

Catechins are useful as high antioxidants which can be used as antibacterial and antiviral [15]. So that when gambier extract is synthesized with other materials, the composite material is not easily decomposed caused by fungi or bacteria.

Research on the manufacture of gel-type cellulose bacteria (in powder) into nano size with a diameter of 3-5 nm using TEMPO oxidation in aqueous media has been widely carried out. This mixture has the advantages of high aspect ratio and modulus of elasticity [16,17]. In a previous study, a flexible supercapacitor that could be bent was found by mixing bacterial cellulose, TEMPO and polypyrrole gel materials [17].

So that on this occasion what will be examined is a material that has high conductivity and is environmentally friendly made from the basic material of cellulose bacterial pellicle. Cellulose bacterial pellicle in this study was thinly ground nata de coco.

Based on the description, the synthesis of bacterial cellulose, TEMPO, gambier, and polypyrrole materials was carried out. Samples of biocomposite 1, biocomposite 2, and biocomposite 3 will be measured for conductivity values using the four point probe method.

## METHOD

### Material

Tools that used in this study were digital scales (Kenko KK-BL), Teflon, dropper pipette, grinding device (Nagako), beaker (Iwaki), hot plate magnetic stirrer (Daihan scientific, MS-H280-Pro), memmert oven (Mettler Universal Oven), multimeter (Sanwa digital multimeter CD800A), and belt sander.

The materials used in this study were nata de coco, 0.5% NaOH, aquades (H<sub>2</sub>O), TEMPO or Tetramethyl Piperidine 1 oxyl (C<sub>9</sub>H<sub>18</sub>NO) (Sigma-Aldrich, Slovakia), acetone (Andeska laboratory, Indonesia), polypyrrole (Sigma -Aldrich, USA), purified gambier (Andalas Sitawa Fitolab, Indonesia), NaBr (Sigma-Aldrich, USA), and FeCl<sub>3</sub> (Andalas Sitawa Fitolab, Indonesia).

### Preparation of pellicle nata de coco

The nata de coco sheets were cleaned using distilled water. Then the nata de coco was soaked in 5% NaOH solution for 24 hours. After that, the nata de coco was rinsed with distilled water to pH 7. Next, the nata de coco was cut and ground into a size of 10 cm long, 1.5 cm wide and 1.2 mm thick. Then soaked in acetone.



Figure 1. Cellulose Bacterial Pellicle in Acetone

### Preparation of TEMPO Bacteria Cellulose

Prepare the Cellulose Bacterial pellicle in acetone. Then stirred 100 ml of distilled water, 0.016 g TEMPO, 0.1 g NaBr for 30 minutes at 350 rpm. Furthermore, the cellulose bacterial pellicle was added to the solution, then the speed was lowered at 200 rpm for 10 minutes. Next, add 1.64 g of NaClO so that the pH becomes 10, then rotate for 15 minutes at a speed of 200 rpm at a temperature of 70° C. Then the Bacteria Cellulose pellicle is boiled in a solution for 40 minutes at a temperature of 70° C, then cooled. Then 5M HCL was added so that the pH was 7. Finally TEMPO Bacteria Cellulose was dried in an oven at 50°C for 2 hours.



Figure 2. Cellulose bacteria pellicle in TEMPO . solution

### Preparation of Gambier Bacteria Cellulose

Pellicle Bacteria Cellulose and 1 g of gambier extract were boiled in 98 ml of distilled water for 1 hour. Then add 5% NaOH solution until pH 7. The solution is allowed to stand for 24

hours. Then rinsed with distilled water and dried in the oven at 50 °C for 2 hours.

Figure 3. Pellicle Bacteria Cellulose boiled in Gambier extract

**Polypyrrole preparation and pellicle mixing**

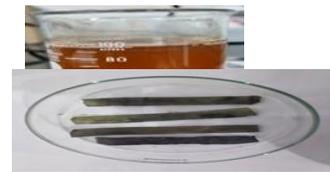
3.66 ml of polypyrrole was mixed with 8 ml of FeCl3 in a petridist. then put in the Cellulose Bacterial pellicle, Cellulose TEMPO Bacteria, and Cellulose Gambier Bacteria for 40 minutes. After that, the pellicle was dried at room temperature.



(a)



(b)



(c)

Figure 5. (a). Samples before being coated, (b). The process of the sample being coated, (c).The process of drying the sample.

**Pellicle conductivity**

Measurement of resistance value using the 4 point probe method. The conductivity value is obtained from

$$\sigma = \frac{1}{4,53 \times t \times R} \tag{1}$$

Where:  $\sigma$  :Conductivity (S/cm)

T : Thickness (cm)

R : Resistance ( $\Omega$ )

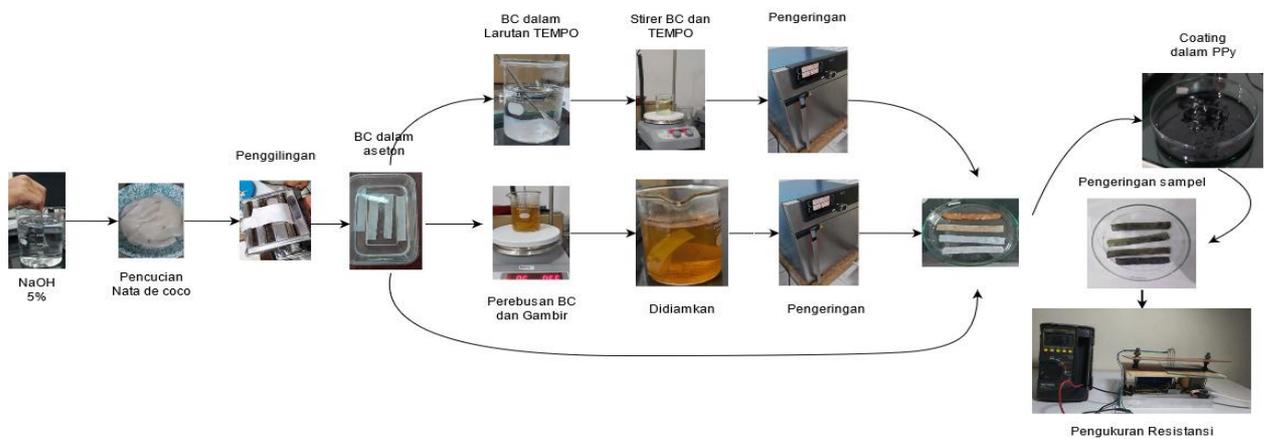


Figure 6. Research procedure

### *The results of the preparation of the pellicle nata de coco*

Cellulose bacteria are soaked in NaOH solution to pH 14. The use of a strong base will kill the bacteria on the cellulose. The residual sediment is cleaned by rinsing using distilled water. The use of aquadest also aims to neutralize the pH and eliminate the sour smell that is still attached to the bacterial cellulose. Because if there are residual components contained in bacterial cellulose, it is estimated that it will block the hydrogen bonds that occur between the cellulose molecular chains which result in a decrease in the strength of the mechanical properties of the bacterial cellulose [18]. Bacterial cellulose was then soaked with acetone and stored in a container at room temperature.

### *The results of the preparation of TEMPO Bacteria Cellulose*

Nanofibers in bacterial cellulose are produced by using TEMPO mixed with NaClO (Sodium hypochlorite) and NaBr (Sodium bromide) at pH 10 and a temperature of 70°C. The results show that the best parameter is the oxidation process in the

**Table 1. Conductivity values of biocomposite 1, biocomposite 2, biocomposite 3, biocomposite 4**

Sample	Resistance ( $\Omega$ )	Thickness(cm)	Conductivity (S/cm)
biocomposite 1	3,530 m	0.25 $\times$ 10 <sup>-2</sup> cm	0.250 $\times$ 10 <sup>-4</sup> S/cm
biocomposite 2	18.63 k	0.54 $\times$ 10 <sup>-2</sup> cm	0.022 $\times$ 10 <sup>-1</sup> S/cm
biocomposite 3	11.88 m	0.21 $\times$ 10 <sup>-2</sup> cm	0.088 $\times$ 10 <sup>-4</sup> S/cm

From table 1, it can be seen that the highest conductivity value is biocomposite 2 and the lowest is biocomposite 3.

The highest biocomposite 2 conductivity value. This is because the absorption of polypyrrole is more by the TEMPO Bacteria Cellulose pellicle compared to other pellicles. More absorption occurs because the TEMPO solution makes the fibers in the Bacteria Cellulose pellicle separate and creates nano-gaps between the fibers. So that when the TEMPO Bacteria Cellulose pellicle is immersed in the Polypyrrole solution, the nano-crevices in the Cellulose Bacteria fiber will be completely filled by the Polypyrrole solution.

Biocomposite 1 sample has a lower conductivity value than biocomposite 2. This is because the Cellulose Bacteria pellicle does not have a nano-gap so that the absorption of polypyrrole does not occur perfectly. Pellicle Bacteria Cellulose has fibers that are close together. When soaked with Polypyrrole solution, the Cellulose Bacteria pellicle covered with Polypyrrole is only on the outside. Due to the nature of the sample of

## RESULTS AND DISCUSSION

manufacture of cellulose nanofibers. The use of TEMPO as a catalyst is very efficient to produce the best nanofibers [19].

### *The results of the preparation of Gambier Bacteria Cellulose*

The purpose of boiling gambier extract is to release catechins from gambier extract cells [20]. In this study, the process of boiling gambier extract and cellulose bacteria was carried out for 1 hour and then deposited for 24 hours. In this process, the catechins from the gambier extract will stick to the cellulose bacteria.

### *Composite conductivity results*

From the test data and calculations using formula (1), the resistance and conductivity values of biocomposite 1, biocomposite 2, and biocomposite 3 are obtained, namely:

biocomposite 2, the conductivity value is lower than that of biocomposite 2.



Figure 6. Biocomposite resistance value measurement 2

The conductivity value of biocomposite 3 is lower than that of biocomposite 1. The conductivity properties of the Gambier Bacteria Cellulose samples were reduced due to the addition of gambier which does not have good electrical conductivity in cellulose bacterial fibers. When the cellulose bacteria are boiled with gambier, the gambier particles will stick to the cellulose bacteria fibers. So that the polypyrrole immersion process does not enter the Gambier Bacteria Cellulose pellicle. However, gambier which is anti-bacterial and fungal causes the

biocomposite 3 sample to be more resistant to fungi and bacteria, so that biocomposite 3 is not easily rotten and moldy.

## CONCLUSIONS

Based on the results of the research that has been done, it can be concluded that:

1. TEMPO's properties are able to separate fibers and cause nano-fiber gaps in cellulose bacteria. This resulted in more polypyrrole covering the nanofibers and bacterial cellulose fibers completely. So that the conductivity value of the cellulose pellicle is high.
2. Gambier which fills the pellicle of cellulose bacteria causes the conductivity value to decrease.
3. The conductivity values of bacterial cellulose film samples with polypyrrole (biocomposite 1) were  $1.178 \times 10^{-3}$  s/cm, the tempo of bacterial cellulose with polypyrrole (biocomposite 2) was  $1.692 \times 10^{-3}$  s/cm, and bacterial cellulose gambier with polypyrrole (biocomposite). 3) ie  $0.9807 \times 10^{-3}$  s/cm. From the measurement using the four point probe method, the highest conductivity value is biocomposite 2, which is  $1,692 \times 10^{-3}$  S/cm.

## ACKNOWLEDGMENTS

On this occasion, I would like to thank my colleagues who have helped to carry out this research. And especially thank you to the Andalas Faculty of engineering who has provided financial assistance, so that this research is carried out to completion.

## REFERENCES

- [1]. Saputra, E.; Sitorus, B. and Harlia, "Synthesis of Polyaniline-Cellulose Composites Using Cellulose Matrix from Oil Palm Empty Fruit Bunches", Tanjungpura University, J. Kimia Equator, vol. 2, no. 1, pp. 58-64, 2013
- [2]. Abe, K., S. Iwamoto and H. Yano, "Obtaining cellulose nanofibers with a uniform width of 15 nm from wood", Biomacromolecules, vol. 8, no. 10, pp. 3276-3278, 2007.
- [3]. D. Sandyakala, "The Effect of Poly (Vynil Pyrrolidone) (PVP) Added in Variation of  $Ca^{2+}$  and  $PO_4^{3-}$  Concentration in Microbial Cellulose-Hydroxyapatite Composite as Scaffold for Bone Healing", Journal of Stem Cell Research and Tissue Engineering, vol. 3, no. 2, 2020
- [4]. Wibowo NA and Isrof, "The In-vivo Potential of Bacterial Cellulose as a Nano-Filler Elastomer Thermoplastics (ETPS)", Perspective, vol. 14, no. 2, pp. 103-112, December 2015.
- [5]. Afrizal, Agung Purwanto, "Utilization of Nata De Coco Bacterial Cellulose as Cu(II) Metal Adsorbant in Water Soluble Systems". Mesomeric Journal vol 1, p. 27-32, 2011.
- [6]. Lusi, Periadnadi, Nurmiati, "The Effect of Sugar Dosage and Addition of Black Tea Extract on Fermentation and Production of Nata De Coco", Journal of Metamorphosis, Vol. IV, no. 1, pp. 126-131, 2017.
- [7]. J. Kim, S. Yun and Z. Ounaies, "Discovery of Cellulose as a Smart Material", Macromolecules, vol. 39, no. 12, pp. 4202-4206, 2006.
- [8]. Pringga Putra. H, "The Effect of Addition of Cinnamon (Cinnamomum Burmanni Bl) and Galangal (Alpinia Galanga L.) Essential Oils on the Characteristics of Biocellulose Film", Pasundan University, 2019
- [9]. Aini NN, Etc., "The Effect of Polymer Type on Reflection Loss in Barium Hexaferrite Matrix Composite (PMC) as Radar Absorbing Material (RAM)", JOURNAL OF ITS ENGINEERING, vol. 5, no. 2, 2016.
- [10]. Wang, H.; Bian, L.; Zhou, P.; Tang, J. and Tang, W., Core-Sheath Structured Bacterial Cellulose/ Polypyrrole Nanocomposites with Excellent Conductivity as Supercapacitors, J. Mater Chem. A., vol. 1, pp. 578-584, 2012.
- [11]. C. Huang et al., "TEMPO-oxidized bacterial cellulose nanofiber membranes as high-performance separators for lithium-ion batteries", Carbohydrate Polymers, vol. 230, p. 115570, 2020.
- [12]. Utami Farida, "Preparation of Nanocellulose from Corn Cobs by Acid Hydrolysis Method at Various Sonication Time Variations", University of Jember, 2018
- [13]. Ayu Fazriyah Rahmi, "Adsorption of Bovine Serum Albumin (Bsa) by Hydrolyzed Cellulose Based on Bagasse (Bagasse)", University of Jember, 2017
- [14]. Kurniatri AA, Etc., "Purification of Catechins from Gambier Extract (Uncaria gambier Roxb.)", Media Litbangkes, vol.29, no. 2, pp. 153-160, June 2019.
- [15]. Silvan JM, Mingo E, Hidalgo M, Pascual-Teresa S, Carrascosa AV, Martinez-Rodriguez AJ. "Antibacterial

- activity of a grape seed extract and its fractions against *Campylobacter* spp. *Food Control*, vol. 29, no. 1, pp. 25-31, 2013.
- [16]. H. Luo et al, "Characterization of TEMPO-oxidized bacterial cellulose scaffolds for tissue engineering applications", *Materials Chemistry and Physics*, vol. 143, no. 1, pp. 373-379, 2013.
- [17]. F. Wang, H. Kim, S. Park, C. Kee, S. Kim and I. Oh, "Bendable and flexible supercapacitor based on polypyrrole-coated bacterial cellulose core-shell composite network", *Composites Science and Technology*, vol. 128, pp. 33-40, 2016.
- [18]. Maulana MI, Etc., "Synthesis and Characterization of Conductive Materials for Polypyrrole (Ppy)/Bacteria Cellulose Composite Films", *JKK*, vol.6, no. 3, pp.11-18, 2017.
- [19]. S. Gea, I. Agrista and C. Zuhra, "Synthesis of Cellulose Nanofibers from Oil Palm Empty Fruit Bunches (Tks) Using the Tetramethyl Piperidine 1 Oxyl (TEMPO) Method", *Talenta Conference Series: Science and Technology (ST)*, vol. 2, no. 1, pp. 22-28, 2019.
- [20]. Irzal, Jasman, and Adri Junil, "Improving the Quality of Boiling Gambier Leaves with the Presto System in Kenagarian Kapuh Pesisir Selatan" *Journal of Mechanical Engineering*, vol. 2, no. 2, pp. 42-47, may 2020.

## AUTHOR(S) BIOGRAPHY



### **Syukri Yunus**

Born in Padang, June 24, 1959. He earned a Bachelor of Engineering (Ir) from the Bandung Institute of Technology (ITB) in 1985. Then a Post graduate at Maryland University in 1989 and then a Master's degree (MSc) from the University of Indonesia in 1990.

Currently serving as a Lecturer in the Department of Electrical Engineering, Faculty of Engineering, and Andalas University.

The author can be contacted at the email address: syukriyunus@eng.unand.ac.id