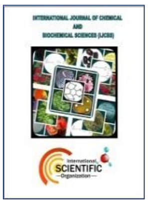
International Journal of Chemical and Biochemical Sciences
(ISSN 2226-9614)

Journal Home page: www.iscientific.org/Journal.html

© International Scientific Organization



Synthesis and characterization of composite membranes based on bacterial cellulose and coral powder

I. Gustian[1]*, A. R. Petinggi[2], E. Maryanti [1], E. Angasa [1], and Nesbah [1]

[1]Department of Chemistry, Faculty of Mathematics and Natural Sciences Universitas Bengkulu, Jalan Raya Kandang Limun, Bengkulu 38371, Indonesia

[2]Bachelor degree Department of Chemistry, Faculty of Mathematics and Natural Sciences Universitas Bengkulu, Jalan Raya Kandang Limun, Bengkulu 38371, Indonesia

Abstract

Preparation of bacterial cellulose and coral powder composite membranes has been carried out. Composite membranes were made by mixing bacterial cellulose with coral powder at different mass ratios at temperatures of 25 ºC, 40 ºC, 60 ºC and At 80 ºC, the composite membrane was evaluated for Fourier Transformation of Infrared, X-Ray Diffraction, degree of swelling, proton conductivity and methanol permeability. From the FT-IR analysis it was found that in the absorption area around 1461.385 – 911cm-1 showing the Si-O-Si stretching functional group, and in the absorption area around 3000 - 3500 cm-1 showing the (O-H) group which shows the semicrystalline composite membrane for each mass variation, according to the results of X-Ray Diffraction. The maximum proton conductivity has been measured as 3.32×10^{-3} $S.cm^{-1}$ at 60°C, produces a degree of swelling of 31.14% and methanol permeability of 3.72×10^{-9} mol/cm.s for a mass ratio of bacterial cellulose:coral powder composite membrane 4:1.

Keywords: Composite membrane, proton conducting, bacterial cellulose, coral powder

Full length article *Corresponding Author*, e-mail: Irfan.g@unib.ac.id Doi # https://doi.org/10.62877/32-IJCBS-24-25-19-32

1. Introduction

Bacterial cellulose is cellulose produced from fermentation by the bacterium *Acetobacter xylinum*, has a high level of purity compared to plant cellulose and has unique structural and mechanical characteristics, has the properties of good strength, low cost, low density, high toughness, and abundant availability [1-4]. Composite is a new material resulting from a combination of two or more materials which does not have the different formation properties of the materials of which it consists filler (matrix) and reinforcement [5]. One of the matrices that is often used in making composite membranes is bacterial cellulose, because bacterial cellulose is easily degraded by the environment, and has high purity, does not experience corrosion, and has good tensile strength relatively large [6]. Bacterial cellulose is easily obtained from nature and is environmentally friendly, namely coconut water fermented with *Acetobacter xylinum* for 6 days, then the resulting bacterial cellulose is phosphorylated by a soaking process for 4 hours the fermentation process is a process of changing sugar into acid organic [7]. Nutrients that play a role in the manufacture of bacterial cellulose are nutrients that contain glucose, namely coconut water and granulated sugar. Glucose that plays a role in the formation of cellulose is glucose in the β form so that all glucose in the α form will be converted into β form through the isomerase enzyme in the bacterium *Acetobacter xylinum*. The polymer synthesized by *Acetobacter xylinum* has unique properties of high mechanical strength, high water absorption capacity, high crystallinity, and very fine and pure fiber structure networks [8], and physicochemical properties. In static culture, cellulose accumulates on a liquid nutrient surface in a gelatinous form, which is called a pellicle or gel. The activity of *Acetobacter xylinum* bacteria with nutrients present in liquid media is the process of pellicle formation. Acetobacter xylinum is a bacterium that produces cellulose, so the nutrient that plays a role is glucose.

Corals are also called stony corals, namely animals from the Ordo *Scleractinia*, capable of secreting CaCO3. Corals belong to the *Anthozoa* class namely members of the Ppylum *Coelenterata* which only have the polyp stage. In the process of forming coral reefs, coral stones (*Scleratina*) are the most important constituent or reef-building coral animal. Stone is a solid, hard and durable substance. Characteristics of some types of stone vary depending on the conditions and how they were formed. One type of rock is coral which has a main mineral composition aragonite (CaCO3), the mineral aragonite is metastable so it can be causing changes to other, more stable forms [9].

Various efforts have been made in order to provide energy that must be able to replace fossil energy sources. The fuel cell is one of the most profitable solutions. The energy produced is quite clean and very efficient, while the processes that occur in fuel cells are based on oxidation-reduction chemical reactions. Requires a hydrophilic proton-conducting membrane on the main chain. Polymers such as cellulose are thought to facilitate the movement of protons, on the other hand, it is possible that cellulose-based membranes can also absorb many water molecules due to the large number of hydrophilic groups in this material. This can cause swelling of the membrane resulting in decreased proton conductivity. Given what has been said so far, it is conceivable to add more inorganic compounds to the membrane to allow proton conduction. for the formation of Bronsted acid-base pairs between two macromolecules [10], the membrane will be able to function as a fuel cell that can operate at high operating temperatures and low humidity levels. In this report, a proton-conducting composite membrane based on bacterial cellulose and coral powder has been carried out with the aim of producing high proton conductivity and other good and superior physicochemical properties.

2. Materials and methods

The samples in this study were samples of coral taken from the coast of Bengkulu City. As for the coconut water taken from the modern traditional market in Bengkulu City, sodium hydroxide, Hydrogen Chloride, ammonium sulphate and *Acetobacter xylinum*.

2.1. Preparation

Bacterial cellulose were made based on previous research [3], coral powder preparation begins by crushing the stone into small sizes of 1-2 cm and then washing it thoroughly using running water, then drying the sample in the sun and placing it on a container, then placing it in the oven at 100 ^oC for 3 hours until the sample is dry. The dry stones were then pulverized using a mortar pestle and then sieved using 100 mesh sieves. After that, 50 grams of coral powder was dissolved in 250 mL of 4 M HCl at 70 ^oC until dissolved and stirred using a magnetic stirrer. 100 mL of 2 M NaOH solution was added to neutralize the solution to pH 7 at 70 ^oC until a precipitate formed and the water content reduced, then centrifuged to separate the remaining solids and liquid, then heated again at 70 ^oC until dry and pulverized using a mortar and pestle. Preparation of composite membranes, bacterial cellulose was blended until smooth to become mush and filtered, then bacterial cellulose membranes with a mass of 5 grams were casted. After that, 0.5 grams, 0.7 gram, and 1 gram of coral powder were weighed. Then added coral powder with bacterial cellulose with a mass ratio of bacterial cellulose and coral powder was 4.5:0.5, 4.3:0.7, 4:1 then stirred until well blended and casting in a petri dish and heated on a hotplate with a heating temperature of 30 ^oC.

2.2. Characterization

On a Bruker Alpha-P (Wismar, Germany), Fourier Transform InfraRed (FTIR) spectra were captured in the attenuated total reflectance (ATR) band of 4000-400 cm^{-1}. Utilizing an X-ray diffractometer (Rigaku D-MAX2200, Japan) and Cu Ka ($\lambda = 1.5406$ Å) radiation spanning the range 2 between 0° and 100°, the X-ray powder diffraction (XRD) investigation was carried out. IM 3590 Chemical Impedance Analyzer HIOKI was used to evaluate the membrane's conductivity at 1 kHz, 0.05 Volt, and temperatures between 25 °C and 80 °C.

.

3. Results and Discussions

The composite membrane and the bacterial cellulose membrane are depicted in Figure 1. Composites have been made by combining coral powder with bacterial cellulose. The mass ratios of cellulose-coral used to make membranes are 4.5:0.5, 4.3:0.7 and 4:1. Determine how the mass affects the properties of the resulting proton-conducting composite membrane, the mass is varied.

Figure 2 shows that there is a typical peak at wave number 3341 cm^{-1} (group O-H) as further absorption from (group O-H) appears at 1650 cm^{-1}, at wave number 1026-1206 cm^{-1} (group C-O stretching for C-O-C and C-O-H). The absorption of this functional group indicates the presence of glycosidic bonds in the cellulose ring [11], namely there are (functional groups O-H and C-O) at absorption wave numbers around 3284 cm^{-1} and 1062 cm^{-1} in bacterial cellulose compounds. Wave number 1576 cm^{-1} shows (functional group C-C), which indicates the presence of a six circular cyclic ring of glucose monomer. From Figure 3 and Figure 4 shows that the typical absorption peaks of coral powder are at wavelengths 1461 cm^{-1} and 911 cm^{-1} which are Si–O–Si stretching groups, indicated as mono and bidental carbonate ligands [12]. Bidental carbonates show absorption peaks at 1420 cm-1 and 875 cm^{-1} [13], and at (C=O group) in the region of wavelength 2126 cm^{-1} it also shows the presence carbonate absorption. Bacterial cellulose composite membrane and coral powder all composite membranes show the same spectrum but it can be seen that the addition of coral at wave numbers 1400 cm^{-1} - 1500 cm^{-1} causes (C–H groups) in bacterial cellulose to react with the ($Si–O–CH_3$) groups to form new clusters with the addition of coral so that it becomes ($Si–CH_3$). vibration shift due to the reaction that occurs.

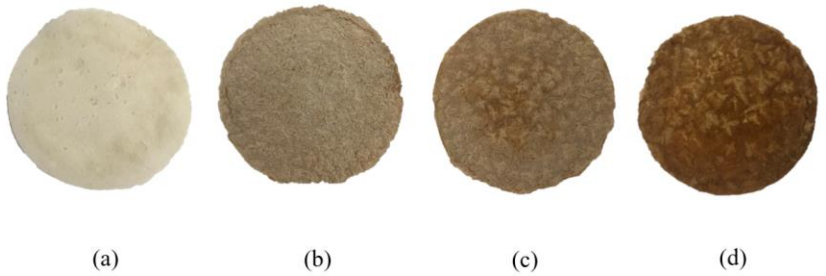


Figure 1: (a) The bacterial cellulose membrane and the composite membranes; (b) 4.5:0.5, (c) 4.3:0.7, (d) 4:1.

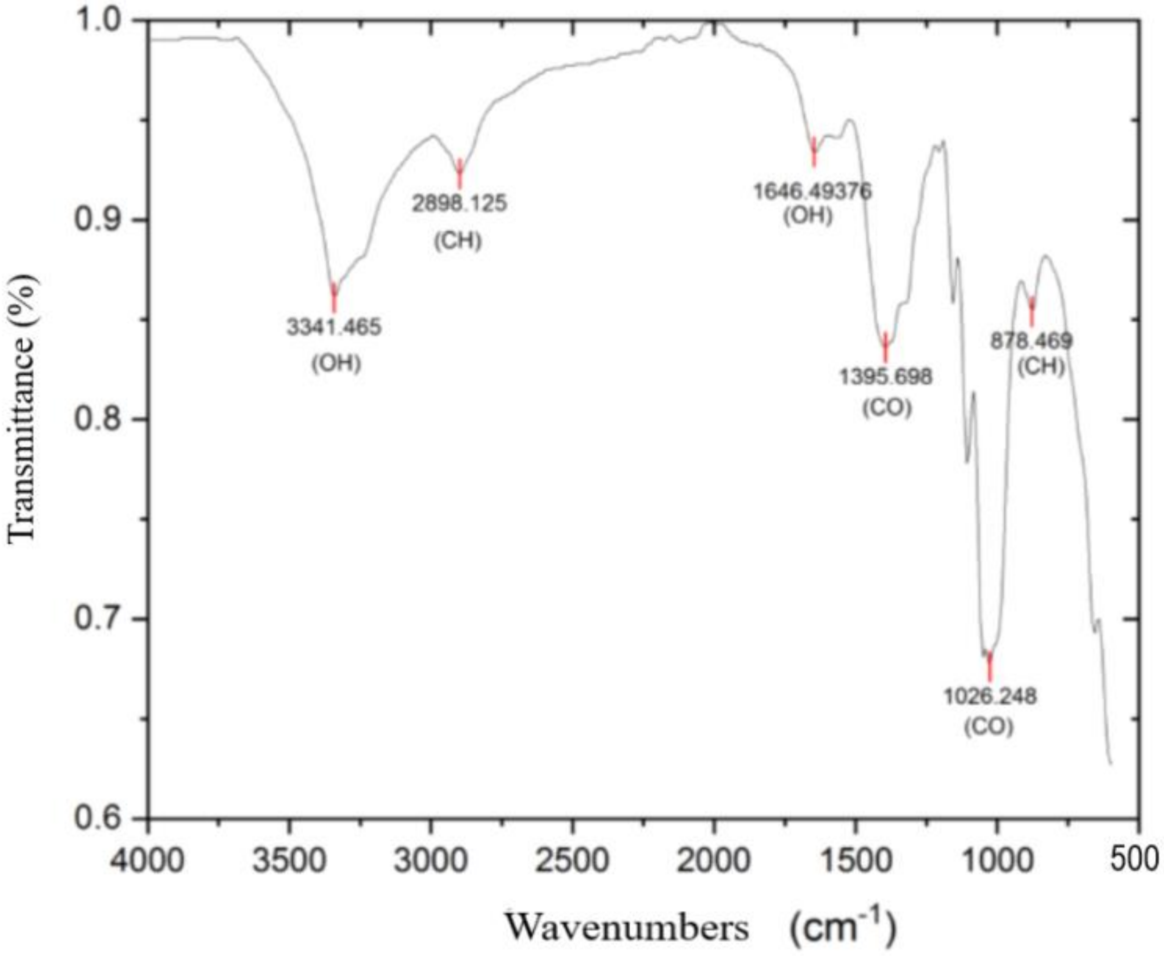


Figure 2: FTIR Spectrum of the bacterial cellulose membrane.

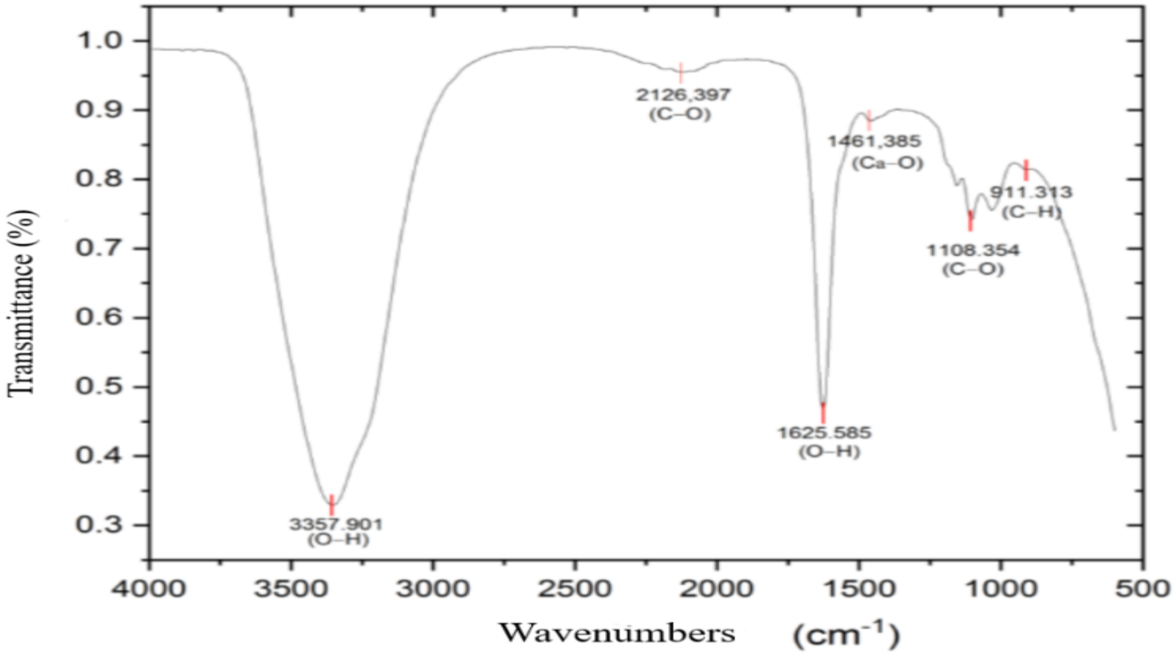


Figure 3: FTIR Spectrum of the coral powder.

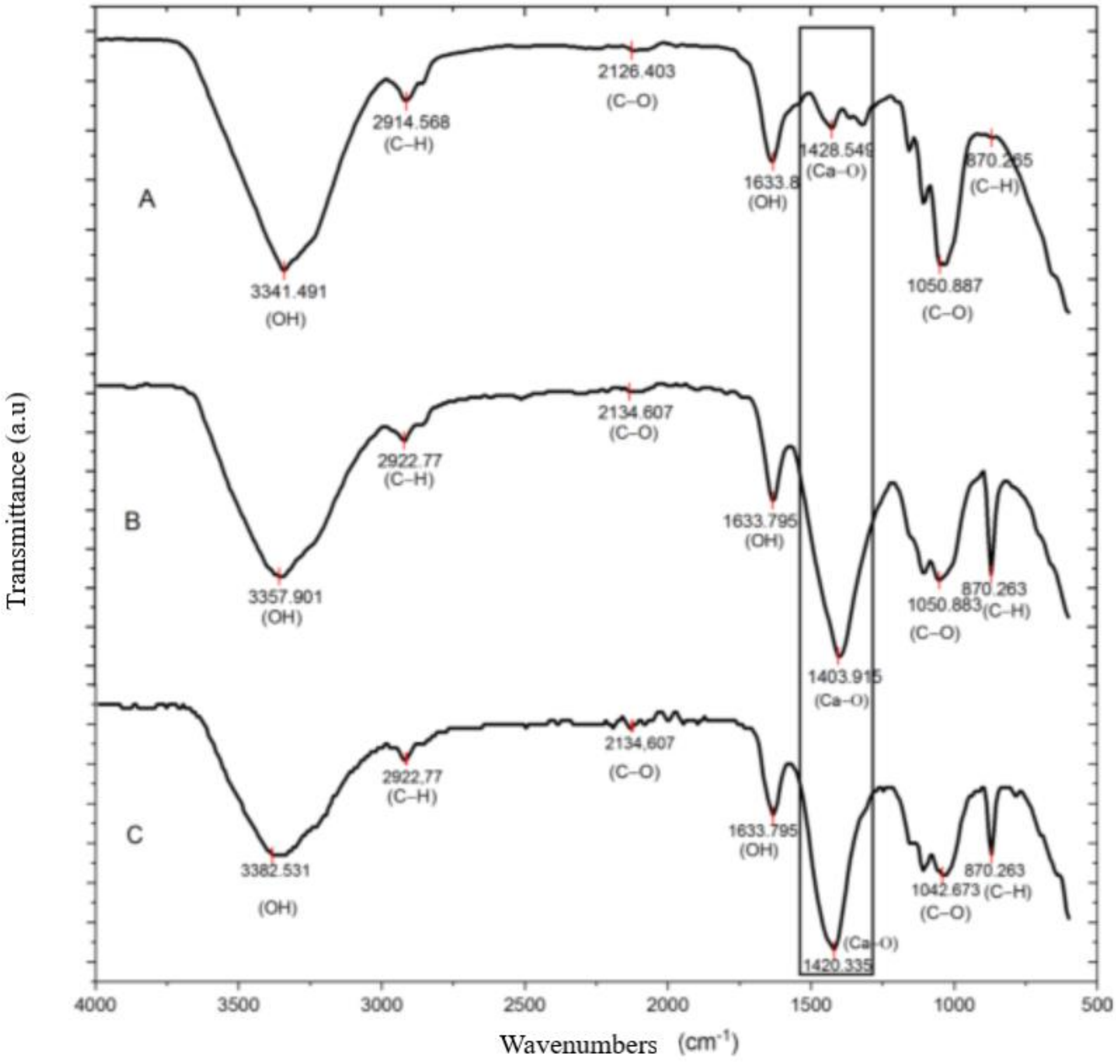


Figure 4: FTIR Spectra of the composite membranes; (A) 4.5:0.5, (B) 4.3:0.7, (C) 4:1.

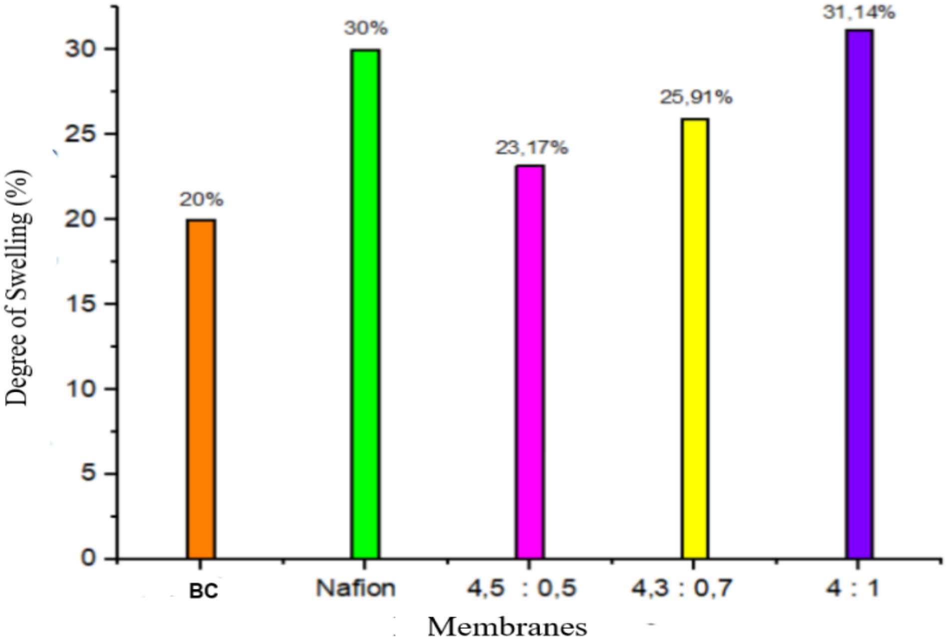


Figure 5: Degree of swelling bacterial cellulose (BC), Nafion, and the composite membranes; 4.5:0.5, 4.3:0.7, 4:1.

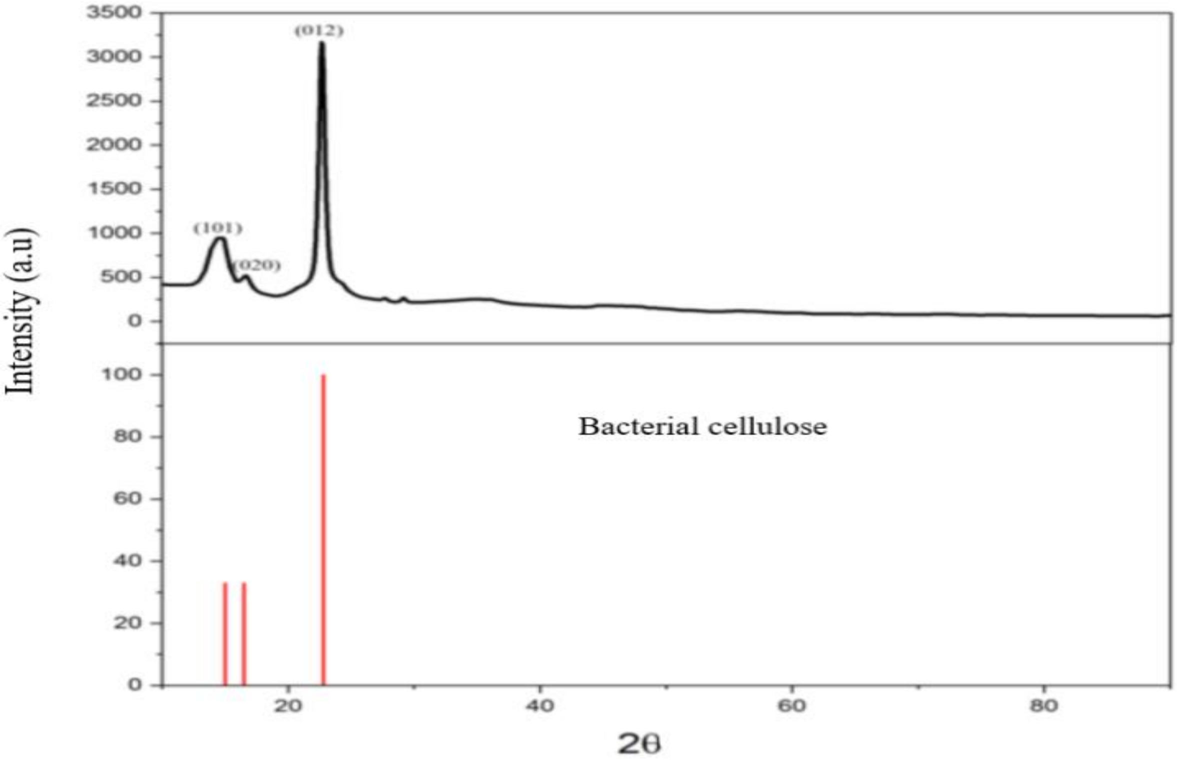


Figure 6: XRD diffractogram of bacterial cellulose.

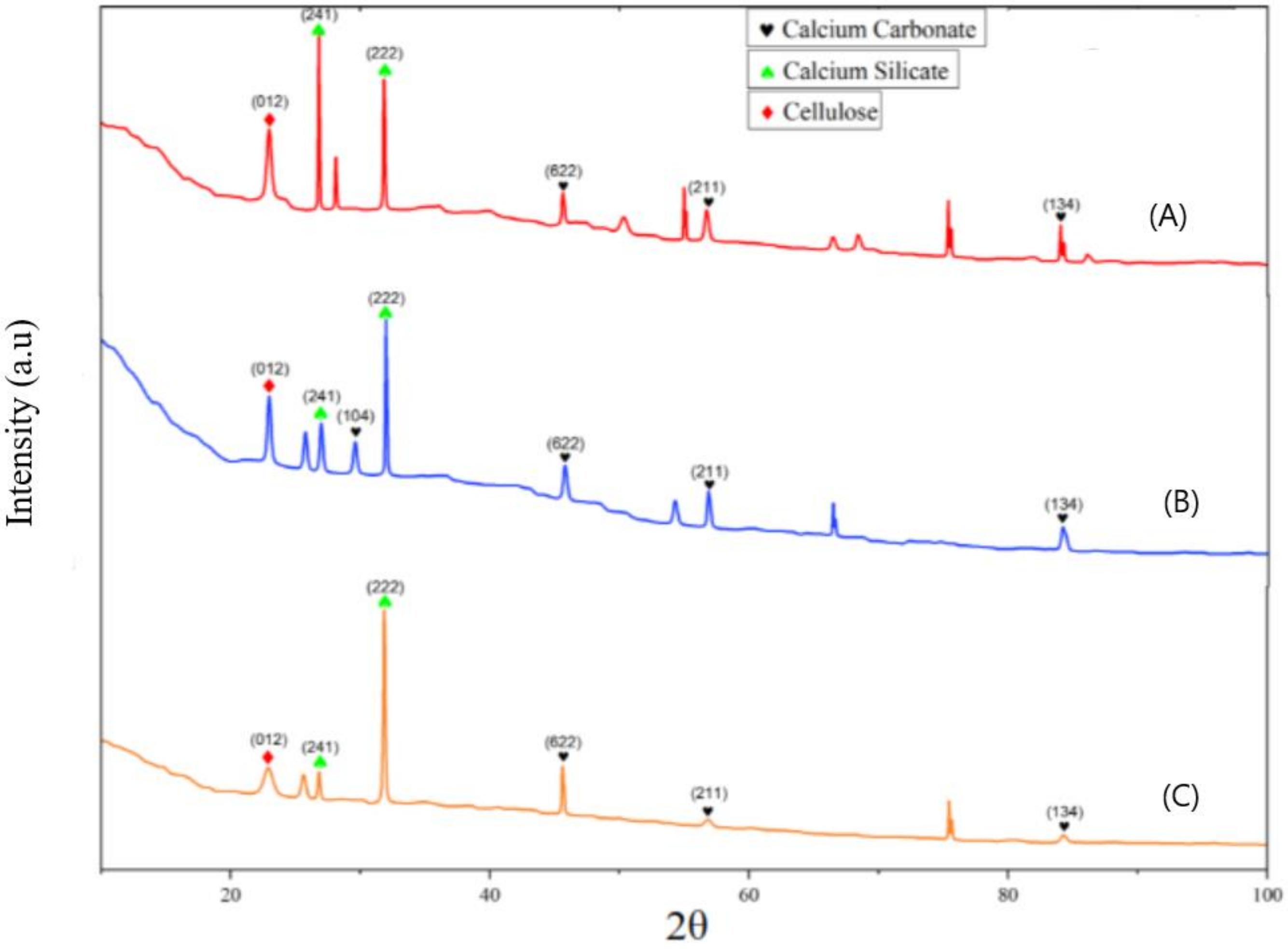


Figure 7: XRD Diffractogram of bacterial cellulose composite membranes: powder coral (A) (4.5:0.5), (B) (4.3:0.7), (C) (4:1).

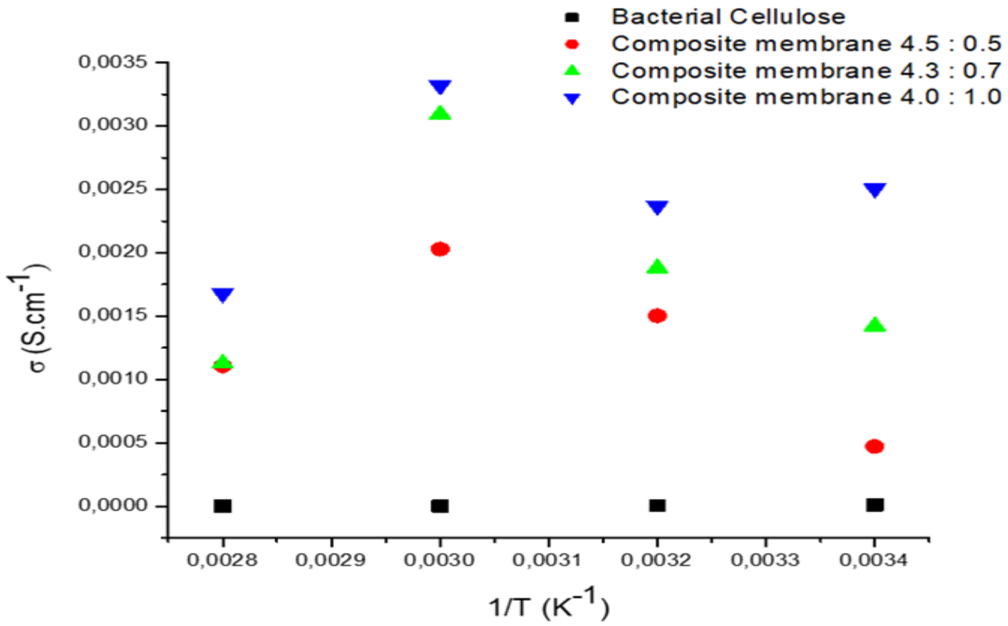


Figure 8: The proton conductivity of bacterial cellulose membranes and composite membranes.

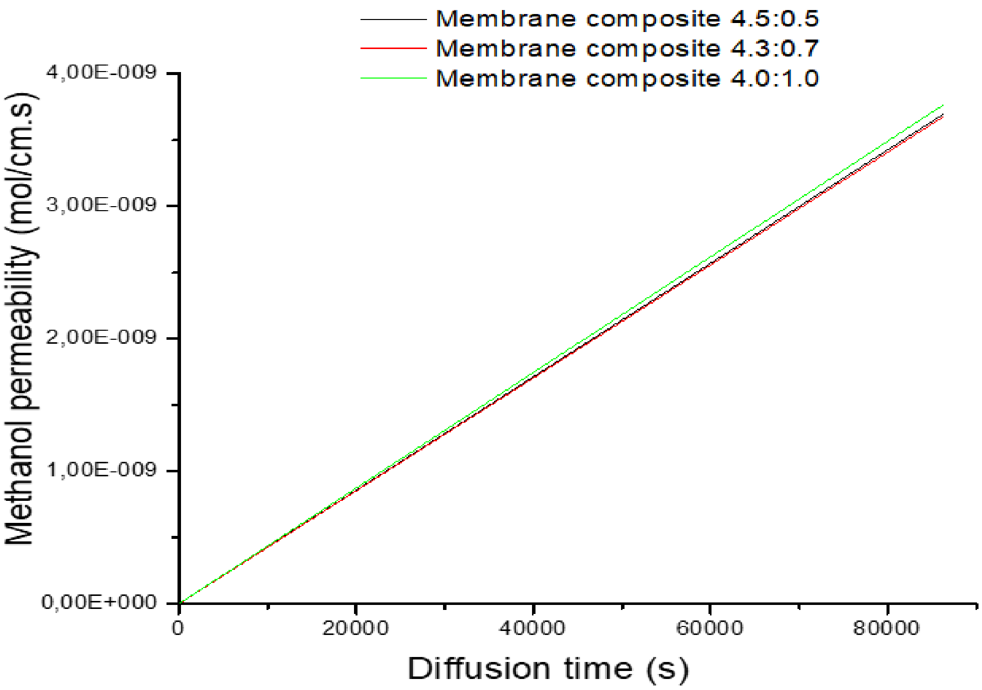


Figure 9: Methanol permeability composite membranes.

From Figure 5 shown that the degree of swelling of pure bacterial cellulose is 20%. the degree of swelling of bacterial cellulose ranges from 17% -36%. The value of the degree of swelling of bacterial cellulose and coral powder after being composited was at a mass ratio of 4.5:0.5 which was 23.17%, at a ratio of 4.3:0.7 it was 25.91%, and at a ratio of 4:1 it was 31.14%. As the amount of coral powder increases, the degree of swelling also increases. This is because coral powder has hygroscopic properties or is able to bind water molecules so that water absorption on the membrane increases. A highwater content in the membrane will facilitate proton transfer, although high water uptake is necessary to obtain high proton conductivity. In general, too highwater uptake can lead to excessive swelling of the membrane and eventual destruction of the membrane. This pattern is almost similar to the degree of swelling of composite membranes based on bacterial cellulose and human nail keratin [1]. Based on the diffraction pattern of bacterial cellulose at Figure 6, there are 2 diffraction peaks found in bacterial cellulose, namely at diffraction $2\theta = 14.4771^{o}, 22.782^{o}$. From Figure 7 has shown diffraction pattern of coral powder that have become composite membranes, there are 4 diffraction peaks, namely at an angle of $2\theta = 32.533^{o}, 46.085^{o}, 56.8671^{o}, 84.3131\ ^{o}$ at the diffraction peaks of 32.533°, 46.085°, and 56.8671° is a calcite diffractogram of the Ca content in the coral. composite of bacterial cellulose and coral powder (4 : 1) has a peak with the highest intensity which is 595.281 at an angle of 31.8451° (4.3 : 0.7) has a peak with the highest intensity experienced by the membrane which is 368.0891 at an angle of 31, 9571° when the ratio of bacterial cellulose was 4.3 : 0.7, the intensity decreased and formed a crystalline phase of $CaCO_3$ with an intensity of 210.6709 at an angle of 29.5911° the $CaCO_3$ angle ranged between 27-29° [14]. For composite membrane 4.5:0.5 showed crystalline peak decreased with an intensity of up to 368.0891 at an angle of 31.9571° and a shift in intensity occurred at an angle of 29.5911° to 239.4671°. Figure 8 showed the proton conductivity of the composite membrane, the highest proton conductivity composite membrane at a temperature of 60 °C is $3.32 \times 10^{-3\ S}$/cm. This matter because the more coral powder added, the higher proton conductivity will increase as seen in Figure 6. This is because the content of coral contains SiO compounds. According to previous reports from other researchers stated that one of the compounds that could increases the proton conductivity of a membrane, namely SiO [15]. The results obtained are the highest conductivity and temperature resistance values good for use in making coral powder and bacterial cellulose composites namely at a temperature of 60°C. The addition of coral powder will increase resistance of cellulose membranes which has been described in previous research that bacterial cellulose only survives at a temperature of 25 °C and at a certain time The addition of coral powder increases heat resistance up to 60°C. This is in line with previous research [16], efforts to increase the proton conductivity and temperature resistance high operation, namely by adding inorganic materials as fillers. However, the proton conductivity showed a decrease which is quite drastic at a temperature of 80 °C. This is because when the water temperature increases on the membrane will evaporate, so that the mobility of the protons will decrease and decreases its proton conductivity. From Figure 9 shown that the measurements methanol permeability with a permeability that is a ratio of 4:1 of 3.72×10^{-9} mol/cm.s and at a ratio of 4.3:0.7 has a permeability 3.6×10^{-9} mol/cm.s and for composite membranes 4.5 : 0.5 has permeability of 3.62×10^{-9} mol/cm.s it can be seen that there is more powderIf coral rocks are added, the permeability will be higher because methanol is more difficult to diffuse if stone dust is added coral. all composite membranes of bacterial cellulose and coral powder shows a higher methanol permeability value than the membrane commercial Nafion 112 which has a methanol permeability value of 1.89 × 10^{-9} mol/cm.s [17].

4. Conclusions

Results of characterization of proton-conducting composite membranes from bacterial cellulose and coral powder, namely the degree swelling of bacterial cellulose was 20% while on the membrane composite have obtained of the degree of swelling for each variation in cellulose mass bacterial cellulose and coral powder show effective for applied to fuel cells, namely 23.17%, 25.91% and 31.14%. The highest proton conductivity for bacterial cellulose is 1.07 × 10^{-5} S/cm at a temperature of 25 ºC and the highest composite membrane in the variation bacterial cellulose:coral powder 4:1 amounting to 3.32 × 10^{-5} S/cm at a temperature of 60 ºC and methanol permeability of 3.72 × 10^{-9} mol/cm.s. The XRD results of bacterial cellulose show 14.4771° and 22.782°, and in coral powder in the 2θ angle area between 32.533° to 56.8671°, the composite membrane is semi-crystalline at each increase in mass.

Acknowledgement

The author would like to thank the Bengkulu University, laboratory staff who have facilitated and helped during the author's research process.

Conflict of Interest

The author reports no conflicts of interest in this work.

Ethical statement

None.

Availability of data and material

We declare that the submitted manuscript is our work, which has not been published before and is not currently being considered for publication elsewhere.

Code availability

Not applicable.

Consent to participate

All authors participated in this research study.

Consent for publication

All authors submitted consent to publish this article research in IJCBS.

References

[1] I. Gustian, A. Simalango, D.A. Triawan, A. M. Hadiputranto, Asdim. (2023). Synthesis and characterization of proton-conducting membranes based on bacterial cellulose and human nail keratin. E-Polymers. 23(1): 41-48.

[2] I. Gustian, Ghufira, A. M. Hadiputranto. E. Angasa, W. Triyono. (2016). Synthesis of proton conductive membrane based on acid-base complex pair: bacterial cellulose and benzotriazole. Rasayan Journal of Chemistry. 9(3): 431-437.

[3] N. Sawitri, Ghufira, D. Fitriani, D.A Triawan, I. Gustian. (2021). Synthesis of proton-conducting membranes based on sulfonated polystyrene and bacterial cellulose. Journal of Physics: Conference Series. 1940(1): 012041.

[4] I. Gustian, E.Widiyati, D. Fitriani, D.A Triawan. (2022). Synthesis and characterization of proton-conducting membranes based on bacterial cellulose and human hair keratin. Rasayan Journal of Chemistry. Special Issue: 59-64.

[5] Gibson, R.F. 1994. Principles of composite material mechanics. New York: Mc Graw Hill.

[6] Darmansyah. 2010. Evaluasi sifat fisik dan sifatt mekanik material komposit serat/resin berbahan dasar serat nata de coco dengan penambahan nanofiller. Tesis. Depok: Fakultas Teknik UI.

[7] A. Mani. (2018). Food preservation by fermentation and fermented food products. International Journal of Academic Research and Development, 1 (Special Issue): 51-57.

[8] S. M. Mohammad, N.A. Rahman, M.S. Khalil, S.R.S. Abdullah. (2014). An overview of biocellulose production using *Acetobacter xylinum* culture. Advences in Biological Research. 8(6): 307-313.

[9] B. D. Erlangga, M. Dedi, Y.C. (2016). Analisis petrograf dan X Ray Diffraction untuk deteksi kalsit non destruktif dari fosil karang porites endapan terumbu kuarter Kendari, Sulawesi Tenggara. Jurnal Riset Geologi dan Pertambangan. 26(1): 15-21.

[10] I. Gustian, S.U. Celik, A. Zainuddin, A.Bozkurt, R.E, Siregar. (2014). Synthesis of polymer electrolyte membrane based on acid-base complex pair and its characteristics. Journal of Mathematical and Fundamental Sciences. 46(1): 50-61.

[11] M. Lindu, P. Tita, I. Erna. (2010). Sintesis dan Karakterisasi Selulosa Asetat dari Nata De Coco sebagai bahan baku membran Ultrafilterasi. Jurnal Sains Materi Indonesia.12(1): 17-23.

[12] Y. Tang, M. Meng, J. Zhang, L, Yong. (2011). Efficient preparation of biodiesel from rapeseed oil over modified CaO. Applied Energy, 88(8): 2735-2739.

[13] A.C.A. Rubio, J.S. Gonzalez, J.M.M. Robles, R.M. Tost, D.M. Alonso, A.J. Lopez, P.M. Torres. (2010). Heterogeneous transesterification processes by using CaO supported on zinc oxide as basic catalysts. Catalysis Today. 149(3-4): 281-287.

[14] B. Lafuente, R.T. Downs, H. Yang, N.Stone. (2015). 1 The power of databases: The RRUFF project. Colombia: Colombia Univerity.

[15] P. Choi, N.H. Jalani, R. Datta. (2005). Thermodynamics and proton transport in Nafion, proton diffusion mechanisms and conductivity. Journal of the electrochemical society. 152(3): E123-E130.

[16] S. Bose, T. Kuila, T.X. Nguyen, N.H. Kim, K.T. Lau, J.H. Lee. (2011). Polymer membranes for high temperature proton exchange membrane fuel cell. Recent advances and challenges. 36(6): 813-843.

[17] S. Unal, S.U. Celik, A. Ata, A. Bozkurt. (2008). Anhydrous proton conducting membranes for PEM fuel cells based on Nafion/Azole composites. Internastional Journal of Hydrogen Energy. 33(11): 2808-2815.