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The Effect of ZnO Mass Variation on Chitosan/ZnO/Cellulose Acetate Composites from Citronella Waste As A Mask Filter Material

Pengaruh Variasi Massa ZnO Pada Komposit Kitosan/Zno/Selulosa Asetat Dari Limbah Serai Wangi Sebagai Material Filter Masker

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ABSTRACT

Air is an important component that affects human survival, but air quality in Indonesia has greatly decreased due to air pollution. This study used chitosan / ZnO / cellulose acetate composite membranes made from citronella waste as mask filters with ZnO variations of 1%, 2%, and 3%. Composite membranes are made by the phase inversion method and characterized by FTIR, tensile, SEM, and antibacterial tests. Optimum conditions based on the formation of pores measuring 0.17 μm are found in chitosan/ZnO/Cellulose Acetate composite membranes with a variation of 3% ZnO. In addition, this variation also has good mechanical properties, with an elongation value of 2.1177% and an elastic modulus of 6.5560 N/m². Based on antibacterial tests, the composite membrane of the 3% ZnO variation also showed the ability to increase antibacterial activity with moderate antibacterial inhibitory strength.

Keywords: *Composite, Filter Mask, Cellulose Acetate, Chitosan, ZnO*

ABSTRAK

Udara merupakan komponen penting yang mempengaruhi keberlangsungan hidup manusia, namun kualitas udara di Indonesia mengalami banyak penurunan karena pencemaran udara. Pada penelitian ini dilakukan pembuatan membran komposit kitosan/ZnO/selulosa asetat dari limbah serai wangi sebagai filter masker dengan variasi ZnO sebesar 1%, 2%, dan 3%. Membran komposit dibuat dengan metode inversi fasa dan dikarakterisasi FTIR, uji tarik, SEM, dan uji antibakteri. Kondisi optimum yang didasarkan pada terbentuknya pori berukuran 0,17 μm terdapat pada membran komposit kitosan/ZnO/Selulosa Asetat dengan variasi ZnO 3%. Selain itu pada variasi ini juga memiliki sifat mekanik baik, dengan nilai elongasi 2,1177% dan modulus young 6,5560 N/m² Berdasarkan uji antibakteri, membran komposit variasi ZnO 3% juga menunjukkan kemampuan dalam meningkatkan aktivitas antibakteri dengan kekuatan daya hambat antibakteri tergolong sedang.

Keywords: Komposit, Filter Masker, Selulosa Asetat, Kitosan, ZnO

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1. INTRODUCTION

Air pollution in Indonesia has reached alarming levels, ranking fifth in the world for worst air quality. Dust particles, released by human activities and natural sources such as volcanoes, contribute to this pollution and can cause respiratory problems and even chronic diseases if inhaled. To reduce the effects of air pollution, masks are recommended. However, disposable masks made from non-biodegradable materials can cause waste buildup. However, more environmentally friendly cloth masks have lower filtering effectiveness due to their single-layer construction.

The alternative solution in question is a mask filter made of cellulose acetate. Cellulose acetate is an organic ester compound that is odorless, tasteless, non-toxic, and biodegradable. It is commonly used in textiles, plastics, paper coatings, and membrane fibers. The cellulose acetate used in this study comes from citronella waste, a by-product of citronella essential oil production. According to Ververis, *et al*. (2004) citronella waste contains 35,0% cellulose, making it suitable as a raw material for cellulose acetate production.

To improve the mechanical properties of cellulose acetate, the addition of chitosan is needed because according to Afidin (2021), cellulose acetate is easy to absorb water, brittle, and stiff. Chitosan increases the water absorption capacity, brittleness, and stiffness of cellulose acetate. Another additive used in mask filter composite membranes is ZnO (zinc oxide), which provides antibacterial properties. ZnO is known for damaging bacterial cell walls, making it an effective bactericide. Compared to other substances such as CuO and Fe2O3, ZnO has shown the highest antibacterial activity against gram-positive and gram-negative bacteria (Azzam, *et al*., 2012). ZnO can attach to the cell membrane bacteria and has an oxidizing ability that can inhibit the growth of bacteria (Tantini, 2020). The manufacture of mask filters from cellulose acetate with the addition of chitosan has previously been carried out by Zomi (2022) and obtained mask filters that have a good filtering system.

This research aims to develop a chitosan/ZnO/Cellulose Acetate composite from citronella waste as a mask filter with better filtration activity. By utilizing the abundant citronella waste and combining environmentally friendly materials such as cellulose acetate and chitosan, the researchers hope to create a sustainable solution to reduce exposure to air pollution through effective mask filtration.

2. MATERIALS AND METHODS

2.1. Materials

The materials used were citronella waste (leaf part), NaOH, NaOCl, distilled water, acetic anhydride, glacial acetic acid, H_2SO_4, ZnO, acetone, ethanol, pp indicator.

The tools used were beakers, measuring cups, measuring flasks, stirring rods, petri dishes, dropper pipettes, funnels, erlenmeyers, spatulas, pH paper, filter paper, magnetic stirrers, blenders, and scissors. In addition, this study also used analytical scales DENVER Instrument Company, glass plates, THERMOLYNE hotplate stirrer, Alpha II FT-IR Infrared Spectroscopy Spectrometer, and Hitachi Flexsem 1000 Scanning Electron Microscope (SEM).

2.2. Methods

The process of cellulose isolation involves delignification and bleaching. To extract cellulose from dried citronella waste, it is first cut into pieces, blended, and filtered through a sieve. The resulting waste is then delignified using a sodium hydroxide (NaOH) solution and heated for three hours. Next, citronella waste is washed until the pH is neutral, and then it is bleached with a sodium hypochlorite (NaOCl) solution until it turns white. The bleached waste is neutralized with distilled water and dried in an oven. The cellulose obtained from this process is then characterized using Fourier Transform Infrared Spectroscopy (FTIR).

In the cellulose acetate synthesis, the delignified cellulose obtained from citronella waste is mixed with sulfuric acid and glacial acetic acid. Acetylation is carried out by adding acetic anhydride and glacial acetic acid, and the mixture is heated for an hour. Afterwards, glacial acetic acid and sulfuric acid are added to the solution, which is heated for an additional hour. The solution is then transferred to a beaker containing distilled water, filtered, and rinsed with water until the pH is neutral. The cellulose acetate is then dried in an oven. FTIR is used to characterize the cellulose acetate.

To produce the chitosan/ZnO/cellulose acetate composite, cellulose acetate is dissolved in acetone and cast onto a glass plate. The plate is then placed in a coagulation bath filled with water. Chitosan is prepared by dissolving it in acetic acid and adding ZnO solids. This mixture is applied to the dried cellulose acetate, and the same process is repeated for variations with different concentrations of ZnO. The composite membrane is characterized using FTIR, tensile testing, scanning electron microscopy (SEM), and antibacterial testing.

For the antibacterial test of the composite membrane, the inhibition zone method is employed against gram-negative bacteria such as E. coli. The membrane is cut into small circles, placed on the surface of an E. coli culture medium, and incubated. The formation of a clear zone around the membrane indicates antibacterial activity.

3. RESULTS AND DISCUSSION

Composite synthesis was done by mixing cellulose acetate, chitosan, and ZnO. The mass variation of ZnO was 1%, 2%, and 3%. The making of this composite is based on the phase inversion method. Phase inversion changes a polymer from a liquid to a solid phase through a certain controlling mechanism. Cellulose acetate dissolved in acetone is cast on a glass plate and then dipped into a coagulation bath filled with water. This aims to dissolve the acetone because cellulose acetate cannot dissolve in water. The cellulose acetate membrane that has been dried is then coated with chitosan that has been dissolved in 2% acetic acid and mixed with ZnO variations of 1%, 2%, and 3%. The dried composite is a thin white membrane. The composite membrane obtained was then analyzed for FTIR, mechanical properties, membrane surface, and antibacterial properties.

3.1. FTIR analysis of composite membrane chitosan/ZnO/cellulose acetate citronella waste

Chitosan/ZnO/Sellulose acetate waste citronella composite membrane was characterized by FTIR to determine the functional groups contained in the composite membrane. **Figure 1** shows the FTIR spectrum of chitosan/ZnO/cellulose acetate waste citronella composite membrane.

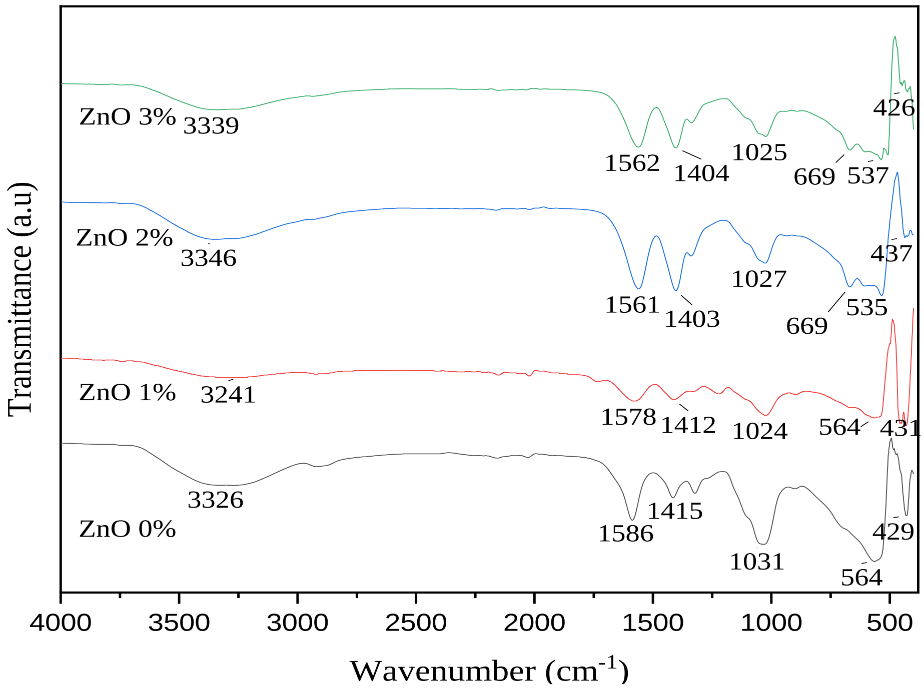

Figure 1. FTIR spectrum of chitosan/ZnO/cellulose acetate composite of citronella waste

The appearance of new absorption in the FTIR spectrum of the composite with the addition of ZnO in the 700-400 cm-1 wavenumber range indicates a ZnO functional group resulting from adding ZnO to the composite (Tantini, 2020). However, similar absorption is found in composites without the addition of ZnO. This can be influenced by chitosan because similar absorption is found in the FTIR of commercial chitosan from the research of Sartika et al. (2016). The absorption indicates that there is still metal content in chitosan. The typical functional group of cellulose acetate, namely C=O, is characterized by absorption in the wavenumber range of 1561-1586 cm-1. The absorption in the wavenumber range of 1024-1031 cm-1 indicates the C-O functional group. The typical chitosan functional group, C-N, is characterized by absorption in the wavenumber range of 1403 - 1415 cm-1. The absorption indicating the -OH functional group at wavenumbers 3750-3000 cm-1 became wider due to the interaction between -OH from cellulose acetate and -NH from chitosan (Rojtica, 2021).

3.2. Analysis of mechanical properties of composite membrane

Chitosan/ZnO/Cellulose acetate waste citronella composite membrane was tested for Tensile strength and then analyzed on the elongation and elastic modulus values to determine the mechanical properties of the composite membrane. Elongation is the maximum strain value of the material when given a force. Meanwhile, elastic modulus (elasticity) is the result of dividing the tensile strength value by the percent elongation (Tantini, 2020). Based on ASTM D838, the elongation value for cellulose acetate-based plastics is 3.10 - 3.50. The results of the composite membrane mechanical properties test can be seen in Table 1.

Table 1 Mechanical properties test results of composite membranes

ZnO variations (%)	Elongations (%)	Elastic modulus (N/m^2)
0	3.1762	6.3029
1	3.1567	7.7188
2	2.4457	5.8997
3	2.1177	6.5560

Adding ZnO to a composite membrane decreases its elongation value, making it more rigid. This is due to the interaction between the polymer matrix and hydroxyl groups on the ZnO particles, which reduces the mobility of the polymer chains. The bond between molecules in the plastic film weakens, causing the pores to open and the composite to become rigid. However, the composite membrane with 1% ZnO variation meets the elongation standard and has potential as raw material for mask filters. The elastic modulus value, which is directly proportional to elongation, is highest in the composite membrane with 1% ZnO mass variation. Adding ZnO reduces the bond density, and agglomeration of the ZnO material can cause a decrease in the elastic modulus value. The 2% ZnO composite has a smaller value than the 3% ZnO composite, possibly due to insufficient bonding between constituent particles.

3.3. Surface analysis of composite membranes

Analysis of the composite membrane surface was carried out using Scanning Electron Spetroscopy (SEM). **Figure 2** shows the SEM results of the composite membrane.

Based on the SEM results, it can be seen that no pores are formed in the chitosan/cellulose acetate composite membrane while in the chitosan/ZnO/cellulose acetate composite with 3% ZnO variation, pores are formed. After analysis, it is known that the pore size formed in the composite membrane is 0.17 µm. This pore size is quite good because it can withstand particles that are harmful to humans which have sizes ranging from ≤ 10 µm and ≤ 2.5 µm and microorganisms measuring 0.1 - 100 µm found in dust. This composite membrane almost meets the ASTM-F2100 mask standard, which can withstand droplets that have a size of 3 µm but is less optimal in holding particles that have a size of 0.1 µm.

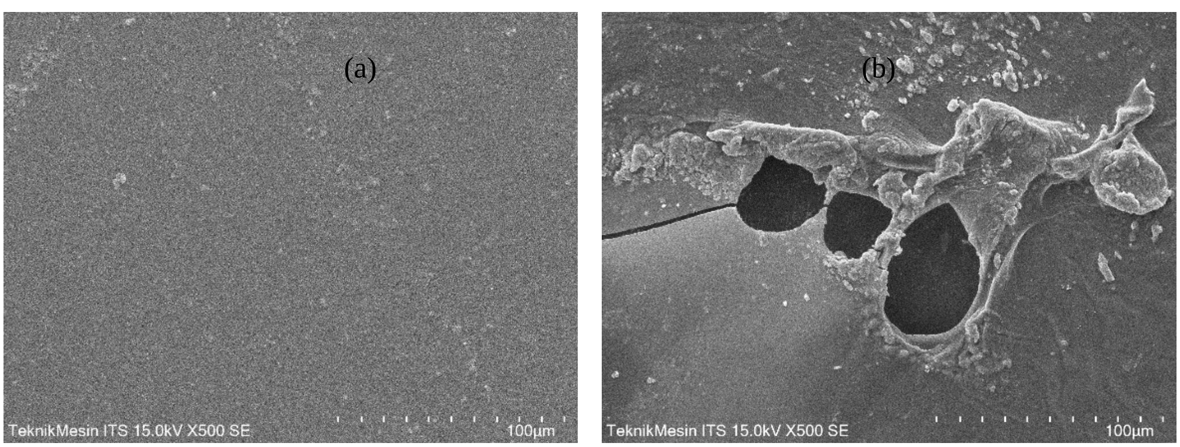

Figure 2. Morphology of composite membrane (a) Chitosan/Cellulose Acetate (b) Chitosan/ZnO/Cellulose Acetate

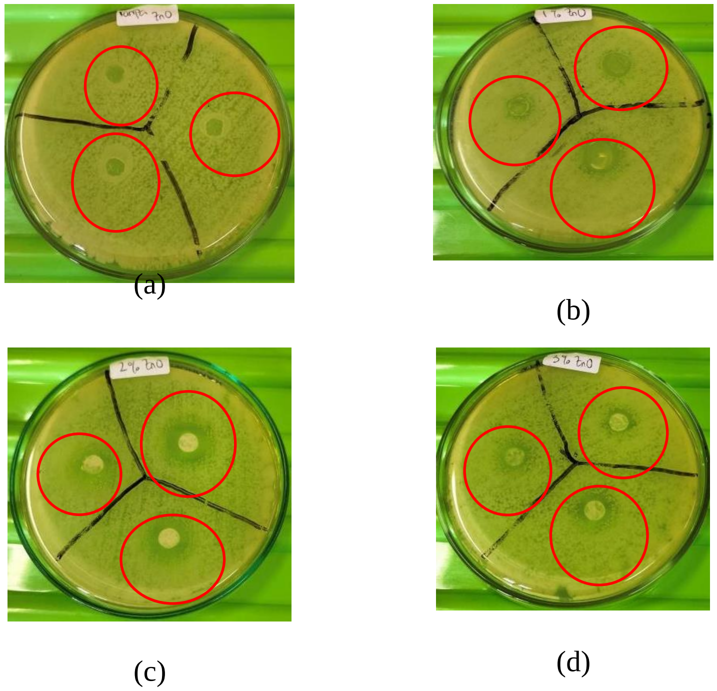

Figure 3. Antibacterial test results of composite membrane (a) 0% ZnO (b) 1% ZnO (c) 2% ZnO (d) 3% ZnO

3.4. Antibacterial properties analysis

The bacteria chosen in this test are E.coli bacteria because they can be carried in the air (Trisno, *et al*., 2019). The zone of inhibition formed on the test sample indicates the presence of antibacterial activity. The positive control used is Erythromycin because it has the ability to kill and inhibit bacterial growth. **Figure 3** shows the zone of inhibition formed in the antibacterial test.

In the picture above, it can be seen that no clear zone is formed on the composite membrane without the addition of ZnO, while the composite membrane with the addition of ZnO forms a clear zone. The clear zone formed is then measured and calculated to determine the strength of antibacterial inhibition. The strength of antibacterial inhibition of the composite membrane can be seen in **Table 2**.

Table 2 Strength of Antibacterial Inhibition

Sample	ZnO variations	Zone of Inhibition Diameter (mm)	Strength of Antibacterial Inhibition
Cellulose Acetate/Chitosan Composite Membrane	ZnO 0%		Weak
Cellulose Acetate/Chitosan/ZnO Composite Membrane	ZnO 1%	5	Medium
	ZnO 2%	9	Medium
	ZnO 3%	6	Medium
Positive Control		30	Very strong

Based on **Table 2**, it is known that the composite membrane without the addition of ZnO has weak antibacterial properties. Liu et al. (2010) said that membranes with the addition of chitosan have less effective antibacterial properties. The addition of ZnO to the composite membrane shows the ability of the membrane to inhibit bacterial growth with moderate inhibition strength. ZnO inhibits bacterial growth by attaching to the surface of the bacterial cell membrane, causing protein denaturation and membrane permeability changes. The bacterial cell wall has a negative charge due to peptidoglycan, composed of long carbohydrate chains intersecting with short amino chains. Zn^{2+} ions from ZnO will bind to the negative charge on the bacterial cell wall, causing the outer membrane of the cell to be damaged. The diameter of the inhibition zone showed an increase along with the addition of ZnO but decreased in the composite membrane with the addition of 3% ZnO. This decrease can be influenced by agglomeration, which causes oxide compounds that inhibit bacterial growth to decrease.

4. CONCLUSIONS

Based on the results of the research that has been done, it can be concluded that based on the pores formed, the optimum condition is found in the chitosan/ZnO/Cellulose Acetate composite membrane with 3% ZnO variation. This is characterized by the presence of pores measuring 0.17 µm. This pore size has met the ASTM-F2100 standard. In addition, this variation also has good mechanical properties, with an elongation value of 2.1177% and a young modulus of 6.5560 N/m2. Based on the antibacterial test, the 3% ZnO variation composite membrane also shows the ability to increase antibacterial activity with an inhibition zone diameter of 6 mm and a moderate antibacterial inhibition strength. This makes the chitosan/ZnO/cellulose acetate composite membrane potentially applied as a mask filter material.

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