

Synthesis of Polyeugenol Sulfonate/Ag⁺ as an Antibacterial Coating on Cotton Fabric

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Abstract: Cotton fabric is widely used in various applications, including the medical field, but its hygroscopic nature makes it prone to microbial contamination. Eugenol a natural phenolic compound extracted from cloves, exhibits potent antibacterial activity due to the ability of hydroxyl group to disrupt microbial membranes. However, its polymerized form, polyeugenol, shows a significantly slower antibacterial response, limiting its immediate effectiveness. In this study, we introduce sulfonate groups to increase the negative charge density of the polymer, potentially improving antibacterial activity. Additionally, Ag⁺ known for its strong antimicrobial effects, was impregnated. Further, synthesized polyeugenol sulfonate/Ag⁺ was applied as an antibacterial coating for cotton fabrics. This material could achieve moderate antibacterial activity with an inhibition zone reaching 7.86 and 8.04 mm against *E. coli* and *S. aureus*, respectively. These improvements in antibacterial activity are promoted to enhance the functionality of cotton fabrics in medical applications, where bacterial resistance is crucial for patient safety.

Keywords: polyeugenol sulfonate; impregnation; silver; spray coating; antibacterial

■ INTRODUCTION

Cotton cloth is a textile material that is widely used for furniture, clothing, and even in the medical field. Cotton fabrics are light, soft, non-sticky, and hygroscopic (very strong at absorbing moisture). This makes cotton cloth an ideal environment for the growth of microorganisms such as mold, *Escherichia coli* and *Staphylococcus aureus* [1]. In the medical field, cotton cloth is widely used in surgery and post-surgery in patients and medical practitioners. If cotton cloth is not treated properly, it can contaminate the cloth and infect the patient and the environment. This is a global challenge that encourages researchers to obtain antibacterial materials so that cotton fabrics can be used in the medical field. The coating of cotton cloth using antimicrobial agents has been shown to kill microorganisms, inhibit microbial growth and prevent bacterial attachment [2]. These properties can be useful in certain circumstances; for example, in the medical field, cotton pads are needed for wound dressings that have antibacterial properties [3].

Eugenol is a natural phenolic essential oil extracted from cloves [4]. The antimicrobial activity of eugenol can be ascribed to the presence of free hydroxyl groups. The hydroxyl group can interfere with the cytoplasmic membrane, thereby increasing nonspecific permeability to various organisms such as *E. coli*, *S. aureus*, and *Pseudomonas aeruginosa* [5-6]. The allylic double bonds in eugenol allow further reactivity or production of functional polymers [7]. Polyeugenol (PE) can be synthesized using a BF₃ catalyst (10 mg/mL), which has antibacterial activity in the slow response category [8]. PE synthesis using H₂SO₄-CH₃COOH catalyst produces high molecular weight polymers with high antibacterial activity, so the molecular weight of the polymer also plays a role in antibacterial activity [9].

Modification of polymers by adding functional groups, such as ionic or ionizable groups in the form of acids, bases, or salts, has the advantage of being able to bind with other materials to increase their function as active ingredients, which leads to improvement of

antibacterial properties. Prasetya et al. [8], modified the polymer using crosslinker DVB and silver nanoparticles to further improve the antibacterial and mechanical properties.

The introduction of sulfonate groups ($-\text{SO}_3\text{H}$) imparts a negative charge to the polymer chain, which enhances its ability to bind with metal ions like silver (Ag^+), known for its potent antibacterial properties [10]. The sulfonate group is known to give a negative charge to the polymer chain so that it can bind to metal materials with good antibacterial properties [11]. Silver is a metal proven to have good antibacterial properties [12]. Ag^+ is unstable and can be reduced to Ag^0 , which limits its practical application. One way to overcome this problem is the protection of silver by a polymer matrix [13]. In this research, studies on the synthesis of PE, polyeugenol sulfonate, and polyeugenol sulfonate- Ag^+ will be carried out which will then be applied to cotton fabrics and tested for their antibacterial activity against *E. coli* and *S. aureus* bacteria.

■ EXPERIMENTAL SECTION

Materials

The materials used in this study were eugenol (Sigma Aldrich), $\text{BF}_3\text{O}(\text{C}_2\text{H}_5)_2$ (Sigma Aldrich), chloroform (Sigma Aldrich), methanol (Sigma Aldrich), Aqua DM (Brataco) Na_2SO_4 anhydrous (Sigma Aldrich), H_2SO_4 (Merck), diethyl ether (Sigma Aldrich), PbSO_4 (Merck), CH_3COOH (Sigma Aldrich), AgNO_3 (Merck), DMSO (Merck), nutrient agar (Millipore-Merck), yeast extract (Millipore-Merck), bacteriological peptone (Oxoid), ciprofloxacin, and cotton cloth.

Instrumentation

The tools used in this study were glasswares, burettes (Pyrex), pH paper (Macherey-Nagel), filter paper (Macherey-Nagel), petri dish, glass spreader, micropipette, ose needle, magnetic hotplate stirrer and reflux, iron spatula, mortar and pestle, stative, analytical balance (Ohaus), oven (Faithful FCD-3000 Serials), scanning electron microscope and energy dispersive X-ray (SEM-EDX, Jeol JSM-6510LA), X-ray diffractometer (XRD, Shimadzu XRD-7000), atomic absorption spectroscopy

(AAS, Perkin Elmer 3110), and gel permeation chromatography (GPC, TOSOH HLC-8320GPC).

Procedure

Synthesis of PE with BF_3 -diethyl ether catalyst

The synthesis of PE with BF_3 -diethyl ether catalyst was done following the procedures previously reported [14]. An amount of 5.8 g eugenol was put into a three-neck flask and 0.25 mL of BF_3 -diethylether was added every 1 h (4 times) while stirring with a stirrer for 12–16 h and stopped by adding 1 mL of methanol. The gel formed was dissolved with chloroform and washed with aqua DM until neutral. After that, anhydrous Na_2SO_4 was added to remove the water. The chloroform was evaporated at room temperature. The precipitate formed was then weighed. The precipitate was analyzed for its polymer structure using FTIR.

Synthesis of PE with H_2SO_4 - CH_3COOH catalyst

Eugenol (5 g) was put into a 250 mL glass beaker. An amount of 1.25 mL catalyst (ratio; 4 mol H_2SO_4 :1 mol CH_3COOH) was gradually added to the cup while stirring (room temperature, 5 min) using a magnetic stirrer. The polymer formation is characterized by the release of thick white smoke and the appearance of purplish black polymer. Then, methanol (1 mL) was added to the cup to stop polymerization. After that, the polymer was precipitated by allowing it to stand at room temperature for 24 h. PE was added to 40 mL of chloroform and stirred until PE was completely dissolved. PE was neutralized and separated with aqua DM to pH 7. The remaining water in the PE is absorbed using anhydrous Na_2SO_4 and then filtered. The solvent was evaporated at room temperature and the precipitate formed was weighed and identified.

Polyeugenol sulfonate synthesis

An amount of 7.5 mL concentrated H_2SO_4 was placed in a three-neck flask and heated at 90 °C. Subsequently, the synthesized PE was gradually added. The mixture was cooled to room temperature, and then 50 mL of cold 2 M H_2SO_4 was added. After that, the solution was filtered and then washed with distilled water until the pH was neutral. The solid obtained is then dried and weighed.

Ag impregnation

AgNO₃ (0.1 g) is put into a 100 mL volumetric flask. Aqua DM was added until it reached the boundary line and then shaken until AgNO₃ was completely dissolved. Poly Eugenol sulfonate was impregnated by contacting it with a solution of AgNO₃ at a ratio of 1:20 (polyeugenol sulfonate:AgNO₃). The mixture was stirred for 24 h in a closed room. After that, the mixture was filtered using filter paper.

Preparation and coating of cotton and polymer fabrics

Cotton cloth is cut into a size of 1.5 cm × 1.5 cm and sterilized by washing in distilled water and 70% alcohol. After that, the cotton cloth was dried at 60 °C for 30 min. Coating of cotton fabrics was carried out using DMSO, ciprofloxacin, Ag, PE-BF₃, PE-H₂SO₄, PE-SO₃H-BF₃, PE-SO₃H-H₂SO₄, PE-SO₃H/Ag-BF₃, PE-SO₃H/Ag-H₂SO₄, which were added 5 drops into the air brush and sprayed with the help of an air compressor at a constant distance from the cotton cloth. Spraying was carried out 3 times. The cloth is dried in an oven at 60 °C for ±1 h, followed by curing at 150 °C for 25 s.

Antibacterial activity testing

Regeneration of bacterial stock on agar media slant.

Test bacteria *E. coli* ATCC 25922 and *S. aureus* were regenerated from stock bacteria with slanted agar media. Agar media was prepared with 25 mg yeast extract, 125 mg peptone, and 1 g of nutrient agar dissolved in 50 mL of distilled water in an Erlenmeyer. After that, the Erlenmeyer was covered with sterile gauze and aluminum foil and then wrapped with plastic wrap to prevent contamination. Erlenmeyer, test tubes, and loop needles will be sterilized by autoclaving at 121 °C for 45 min. The process was then continued in laminar air flow (LAF), as much as 5 mL of each medium was poured into 3 test tubes and allowed to solidify at room temperature, with the position of the tube tilted 30° to the plane. After compaction, the test bacteria were inoculated on the slanted agar media using a sterile loop needle. After that, incubation was carried out in an incubator for 24 h at 37 °C.

Preparation of bacterial inoculum according to McFarland standards. Liquid media was prepared with 25 mg of yeast extract and 125 mg of peptone dissolved in

50 mL of distilled water in an Erlenmeyer. After that, the Erlenmeyer was covered with sterile gauze and aluminum foil and then glued with plastic wrap to prevent contamination. The Erlenmeyer, test tube, and loop needle that will be used are sterilized by autoclaving for 45 min. After that, the bacterial suspension process was carried out in liquid media in an Erlenmeyer flask, which was carried out in the LAF. Incubation was carried out in an incubator shaker using a temperature of 37 °C and absorbance measurements were carried out at a wavelength of 600 nm until turbidity was obtained according to the McFarland standard of 0.5.

Testing the antibacterial activity of materials with the disc diffusion method. The agar media that had been cooled in LAF were inoculated with bacteria, as much as 30 µL of bacterial suspension according to the 0.5 McFarland standard. The disc paper that has been dripped with 10 µL sample is then placed on the surface of the test media, and then an incubation process is carried out for 24 h with observational treatment. After that, the clear zone measurements were taken at the 12th and 24th h to determine the differences in the clear zones formed.

Testing the antibacterial activity of fabrics with the turbidimetric method. The liquid media was prepared by inoculating 50 µL of bacterial suspension according to the 0.5 McFarland standard. Fabric samples measuring 1.5 cm × 1.5 cm were put into the liquid medium and incubated in an incubator shaker for 12 h at 50 rpm. After that, the absorbance was measured at 600 nm with a UV-vis spectrophotometer. The absorbance of bacterial cultures incubated with the treated fabric was measured and compared to cultures incubated with untreated (pristine) fabric to evaluate the antibacterial activity of the polymer-coated fabric.

Testing hydrophilicity of treated fabrics using the sessile drop method. A droplet of distilled water (typically 3–5 µL) was placed on the surface of the fabric using a pipette, and the contact angle formed between the water droplet and the fabric surface. The contact angle was measured from droplet images using image analysis software by drawing a tangent at the droplet–surface interface and determining the angle formed between the droplet edge and the fabric surface.

■ RESULTS AND DISCUSSION

PE Synthesis

The synthesis of PE was carried out with two different catalysts, namely $\text{BF}_3\text{-O}(\text{C}_2\text{H}_5)_2$ and $\text{H}_2\text{SO}_4\text{-CH}_3\text{COOH}$. The synthesis was carried out for 16 h through a cationic addition process, which caused the allyl group in eugenol to undergo an addition reaction. In the initiation step (Fig. 1(a)), the double bond in the eugenol allyl group breaks and forms a carbocation. In the propagation stage (Fig. 1(b)), the carbocation formed will bind to other carbocations that form the polymer chain. The reaction was terminated at the termination stage (Fig. 1(c)) by adding methanol. The polyeugenol that was formed was in the form of a solid gel, which was dissolved in chloroform and neutralized with demineralized water to remove the acid from the remaining catalyst. Anhydrous Na_2SO_4 is used to bind the remaining water.

PE with the catalyst $\text{BF}_3\text{-O}(\text{C}_2\text{H}_5)_2$ is orange with an average molecular weight of 12753 g/mol, an average total molecular weight of 10160 g/mol, and a polydispersity

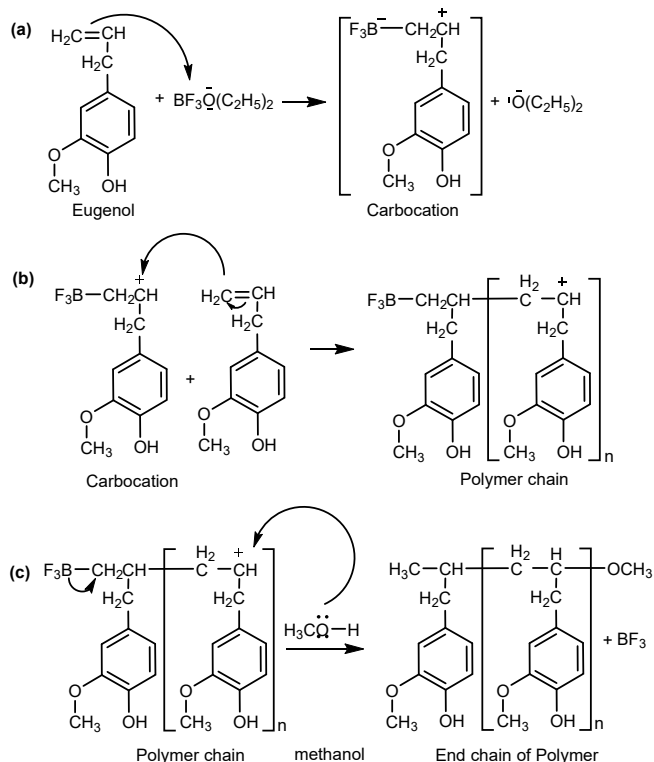


Fig 1. Steps of polymerization: (a) initiation, (b) propagation, (c) termination

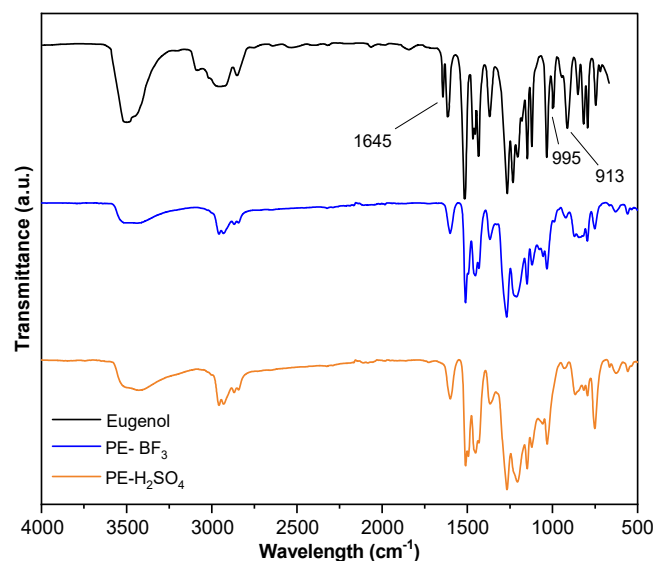


Fig 2. FTIR spectra of eugenol and PE with different catalysts

index 1.25. PE with catalyst $\text{H}_2\text{SO}_4\text{-CH}_3\text{COOH}$ is brown with an average molecular weight of 16983 g/mol, an average total molecular weight of 13389 g/mol, and a polydispersity index 1.27. Both PEs were characterized using FTIR and compared with eugenol in Fig. 2. At 995 and 913 cm^{-1} , the loss of the vinyl group that was previously present in eugenol. The vinyl group has been modified to bind with other eugenols and form PEs.

Polyeugenol Sulfonate Synthesis

Synthesis was carried out by reacting the synthesized PE with concentrated H_2SO_4 . The sulfonate group will enter the meta position of the $-\text{OCH}_3$ group because the $-\text{OH}$ group is an ortho and para directing group. The ortho position on the right and the para position are already filled, so the SO_3^- group will enter the ortho position on the left. Polyeugenol sulfonate with catalyst $\text{H}_2\text{SO}_4\text{-CH}_3\text{COOH}$ (PE- $\text{SO}_3\text{H-H}_2\text{SO}_4$) in the form of black powder with a yield percentage of 80%. The molecular weight obtained was 11943.96 g/mol. In Fig. 3(a), the S=O group at 1026 and 1158 cm^{-1} ; a CS group at 623 cm^{-1} . This proves that polyeugenol was successfully sulfonated. Polyeugenol sulfonate catalyst $\text{BF}_3\text{-O}(\text{C}_2\text{H}_5)_2$ (PE- $\text{SO}_3\text{H-BF}_3$) is in the form of black powder with a yield percentage of 78%. The molecular weight obtained was 10182.77 g/mol. Fig. 3(b) shows S=O groups at 1026 and 1155 cm^{-1} ; a CS groups at 625 cm^{-1} indicate sulfonate groups in polyeugenol.

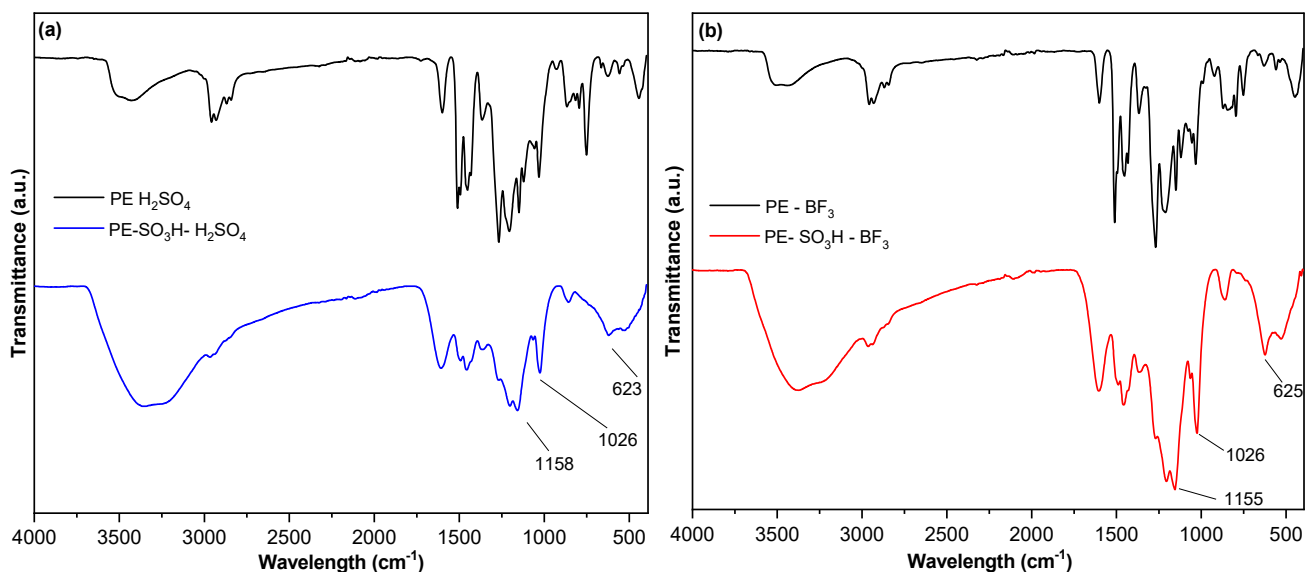


Fig 3. FTIR spectra of (a) PE with the catalyst $\text{H}_2\text{SO}_4\text{-CH}_3\text{COOH}$ and (b) PE with the catalyst $\text{BF}_3\text{-O}(\text{C}_2\text{H}_5)_2$

Polyeugenol Sulfonate Impregnation with Ag^+ Ions

The results of the impregnation of Ag^+ ions with polyeugenol sulfonate can be seen from the XRD analysis (Fig. 4 and Table 1). On the XRD diffractogram Fig. 4, there is a peak similarity between the sample and the Ag standard at 2θ : 38.18° ; 44.44° ; 64.57° ; 77.42° . The peaks between the samples and the appropriate standards indicated that Ag was bound to $\text{PE-SO}_3\text{H}$. The results of the EDX analysis (Table 2) show that the coated fabric contains Ag. To evaluate the interaction between Ag^+ ions with the polymer, the concentration of Ag^+ in solution was measured before and after exposure to the polymer using atomic absorption spectroscopy (AAS). The data shows a significant reduction in Ag^+ concentration after contact with polymer (over 84% in both samples), indicating adsorption of Ag^+ into the polymer matrix and

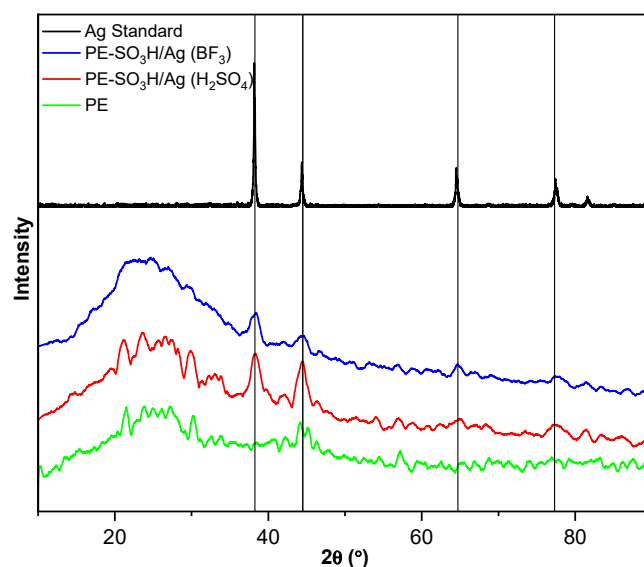


Fig 4. Comparison of the standard XRD diffractograms of Ag, $\text{PE-SO}_3\text{H/Ag}$, and PE

Table 1. $\text{PE-SO}_3\text{H}$ contact result data

Sample	Before contact (ppm)	After contact (ppm)	Adsorbed concentration (ppm)	Adsorbed percentage (%)
$\text{PE-SO}_3\text{H/Ag}^+(\text{BF}_3)$	608.311	84.774	523.537	86.000
$\text{PE-SO}_3\text{H/Ag}^+(\text{H}_2\text{SO}_4)$	608.311	91.706	516.605	84.900

Table 2. Percentage by mass of the elements in the fabric

Composition	Mass percentage (%)		
	Plain clothing	$\text{PE-SO}_3\text{H/Ag-BF}_3$ clothing	$\text{PE-SO}_3\text{H/Ag-H}_2\text{SO}_4$ clothing
C	61.56	61.84	61.28
O	38.44	37.90	38.42
Ag	-	0.27	0.31

suggesting that the polymer has a strong affinity for Ag^+ ions. With an initial concentration of Ag^+ of 608.311 ppm, contacting with $\text{PE-SO}_3\text{H}/\text{Ag}^+$ (BF_3), dropping the concentration to 84.774 ppm, resulting in a percentage of adsorption up to 86%. Similarly, with $\text{PE-SO}_3\text{H}/\text{Ag}^+$ (H_2SO_4), the concentration decreased to 91.706 ppm, resulting in a percentage of adsorption of 84.9%. The high adsorption efficiency suggests an active interaction between Ag^+ ions and the polymer matrix, specifically with the SO_3H group.

Antibacterial Activity Test

Material antibacterial test with disc diffusion method

Antibacterial activity test was carried out with three different concentrations, namely 5.0, 12.5, and 20.0 mg/mL for each sample, 2.5 mg/mL for the positive control, and 1 mg/mL for Ag. The positive control used was ciprofloxacin. The inhibition zone was measured using a digital vernier caliper. Measurements of the clear zones were taken at the 12th and 24th h to determine any differences in zone size over time. Measurement of the inhibition zone in Fig. 5 indicated that the best antibacterial was polyeugenol with a concentration of 20 mg/mL. The greater the concentration of the material, the greater the antibacterial activity of both *E. coli* and *S. aureus* bacteria. This is because the higher the concentration, the more compounds that are able to diffuse into bacteria through the cell wall, so that they have the potential to inhibit bacterial growth. Statistical analysis using one-way ANOVA revealed that the

differences in inhibition zone sizes across the different concentrations were statistically significant ($p < 0.01$), confirming that the increase in inhibition zone size with concentration is not due to random variation.

The diameter of the inhibition zone obtained with *E. coli* bacteria was smaller than that of *S. aureus* bacteria. This is due to the influence of differences in the bacterial cell wall structure in the two bacteria. Gram-negative bacteria (such as *E. coli*) are surrounded by two membranes, where the outer monolayer contains lipopolysaccharide as the main lipid component, making it more difficult to diffuse through the cell wall. Gram-positive bacteria (such as *S. aureus*) have a simpler, single-layered cell wall structure and lower lipid content. Gram-negative bacteria tend to be more resistant to antimicrobial agents than Gram-positive bacteria because of the added protection provided by the outer membrane, apart from this outer membrane [15].

The use of a catalyst affects the molecular weight of the PE formed. Based on the comparison of the inhibition zones in Fig. 6, $\text{PE-H}_2\text{SO}_4$ has higher antibacterial activity than PE-BF_3 . The hydroxyl group ($-\text{OH}$) has an important role in inhibiting bacterial metabolism. This group will interact with proteins in bacteria, disrupting bacterial cell membranes, which leads to increased permeability, leakage of ions and proteins, and cell death [16]. The higher the molecular weight, the longer the monomers that make up the polymer chain. PE with a larger molecular weight will have more hydroxyl groups, so the antibacterial activity is higher.

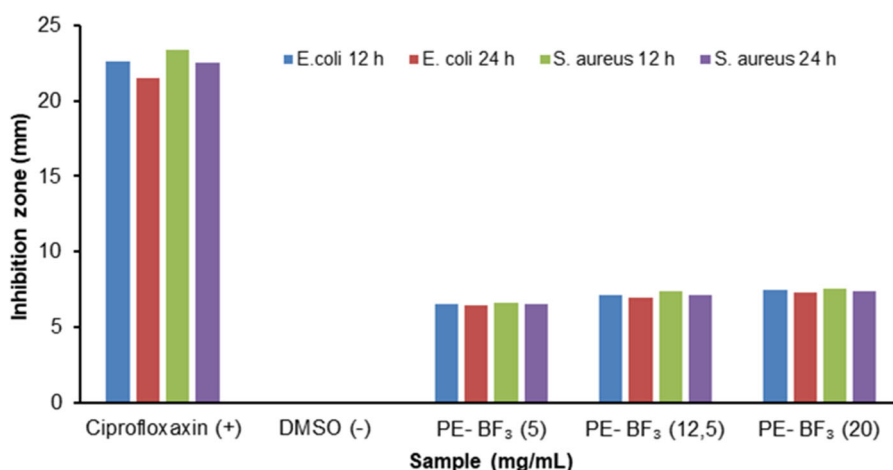


Fig 5. Inhibition zone comparison diagram based on concentration

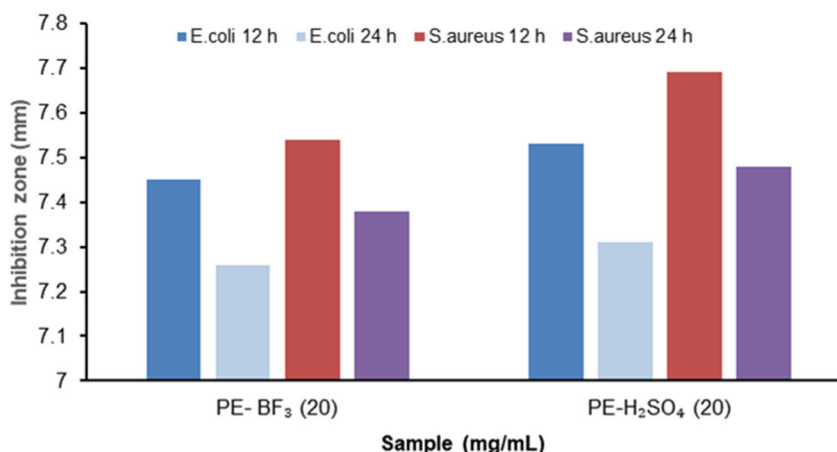


Fig 6. Inhibition zone comparison diagram based on the catalyst

The addition of sulfonate groups to PE increased the inhibition zone formed, as shown in Fig. 7. The sulfonate groups cause the aqueous media in the microbes and/or their suspensions to simultaneously increase the hydration of the polymer and lower the pH. Bacteria tend to maintain a neutral internal state regardless of external pH. Sudden changes in pH increase the pressure on the outer membrane; if the change is drastic enough, it can damage the membrane resulting in enzyme damage, protein denaturation, and microbial death [16]. In Fig. 8 the inhibition zone of PE-SO₃H/Ag⁺ is larger than that of PE-SO₃H in *E. coli* and *S. aureus* bacteria. This indicates that Ag⁺ ions impregnated into the PE matrix have a role in increasing antibacterial activity. Enhanced antibacterial performances of PE-SO₃H/Ag⁺ are likely due to the synergistic effect between both PE-SO₃H and Ag⁺ ion. The presence of a sulfonated group may facilitate the binding of Ag⁺ ions and possibly help gradually release

Ag⁺ ions over time. Negatively charged sulfonated groups may enhance Ag⁺ interaction with the bacterial cell, which improves the impact. Those synergistic effects contribute to the observed increase in inhibition size. The diameter of the resulting inhibition zone was 6–10 mm, so it can be stated that both PE, PE-SO₃H, and PE-SO₃H/Ag⁺ at a concentration of 20 mg/mL have moderate antibacterial activity. In Table 3, comparing this material with previously reported polyeugenol-based material, our material showed improvement against *E. coli* bacteria, which can offer substantial potential for many applications.

Antibacterial activity test of fabric coated materials with the turbidimetric method

The results of the activity test with the turbidimetric method are presented in Table 4. There is an increase in the percentage of inhibition after being coated with the material. Unmodified PE cloth prepared

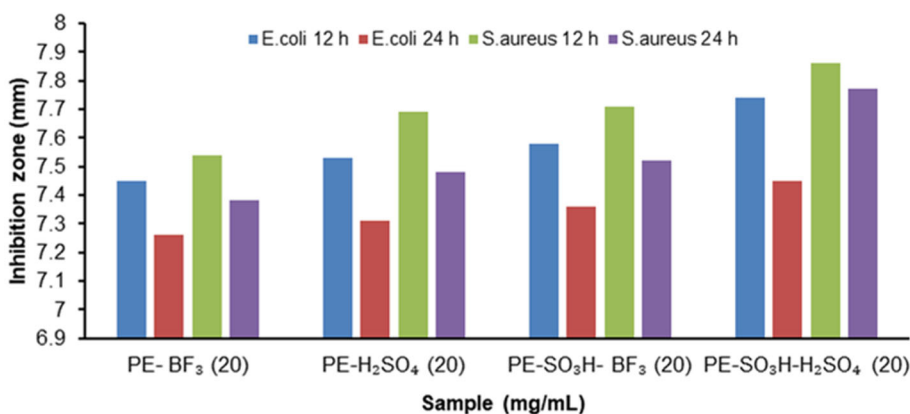


Fig 7. Comparison diagram of PE and PE-SO₃H inhibition zones

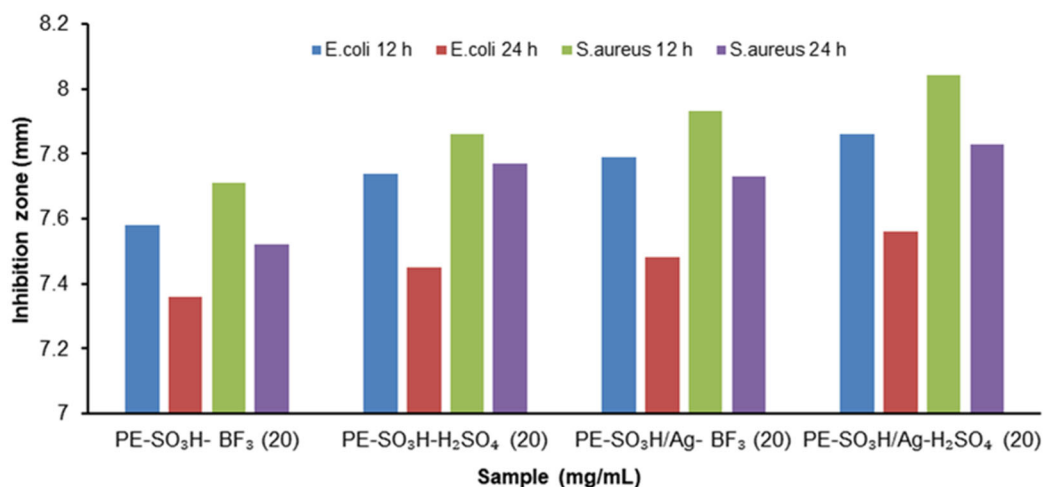


Fig 8. Comparison diagram of PE-SO₃H and PE-SO₃H/Ag⁺ inhibition zones

Table 3. Inhibition zone comparison with polyeugenol-based material

Material	Concentration	Inhibition zone (12 h)		Reference
		<i>E. coli</i>	<i>S. aureus</i>	
Polyeugenol	10 mg/mL	0.35	2.46	[17]
PEDVB	20 mg/mL	0.87	2.27	[8]
PEDVB-SO ₃ H/AgNP	30 ppm	7.50	8.50	[15]
PE-SO ₃ H/Ag-H ₂ SO ₄	20 mg/mL	7.86	8.04	Our work

Table 4. Data on the percent inhibition of coated cotton fabrics

No	Sample	Abs.	%inhibition <i>E. coli</i>	Abs.	%inhibition <i>S. aureus</i>
1	Plain cloths	0.781	0.000	0.713	0.000
2	Cloth + CPX (+)	0.001	99.870	0.002	99.670
3	Cloth + DMSO (-)	0.731	6.440	0.697	2.200
4	Cloth + Ag (1000 ppm)	0.003	99.620	0.013	98.180
5	Cloth + PE-BF ₃	0.709	9.220	0.629	11.740
6	Cloth+ PE-SO ₃ H-BF ₃	0.682	12.670	0.596	16.320
7	Cloth + PE-SO ₃ H/Ag-BF ₃	0.623	20.260	0.556	22.030
8	Cloth + PE-H ₂ SO ₄	0.692	11.390	0.623	12.540
9	Cloth + PE-SO ₃ H-H ₂ SO ₄	0.622	20.350	0.548	23.060
10	Cloth + PE-SO ₃ H/Ag-H ₂ SO ₄	0.535	25.510	0.521	26.940

with BF₃ or H₂SO₄ catalyst reduces the growth of bacteria modestly (~9–12% for *E. coli* and 12–13% for *S. aureus*). Introducing sulfonate groups doubled the antimicrobial activity, pushing the inhibition to ~20%. The PE-SO₃H/Ag⁺-H₂SO₄ coated cloth had an inhibition percentage of 25.51% for *E. coli* bacteria and 26.94% for *S. aureus* bacteria. These results support previous disc diffusion tests and prove that PE-SO₃H/Ag⁺ has the potential to be an antibacterial coating material for cotton fabrics.

Fabric Surface Morphology Analysis

Uncoated cotton fabric (Fig. 9(a)) has a morphological structure with clean bands and no visible coating on its surface. There is a thickening and a layer covering the surface of the cloth on the PE-SO₃H/Ag⁺ coated cloth (Fig. 9(b) and (c)).

Hydrophilicity Test

The purpose of the hydrophilicity test is to determine the level of hydrophilicity of the coated fabric

using the sessile drops method. Fabrics are hydrophobic when they have a contact angle of $> 90^\circ$ whereas fabrics

are hydrophilic when they have a contact angle of $< 90^\circ$ [18]. The data presented in Fig. 10 and Table 5 highlight

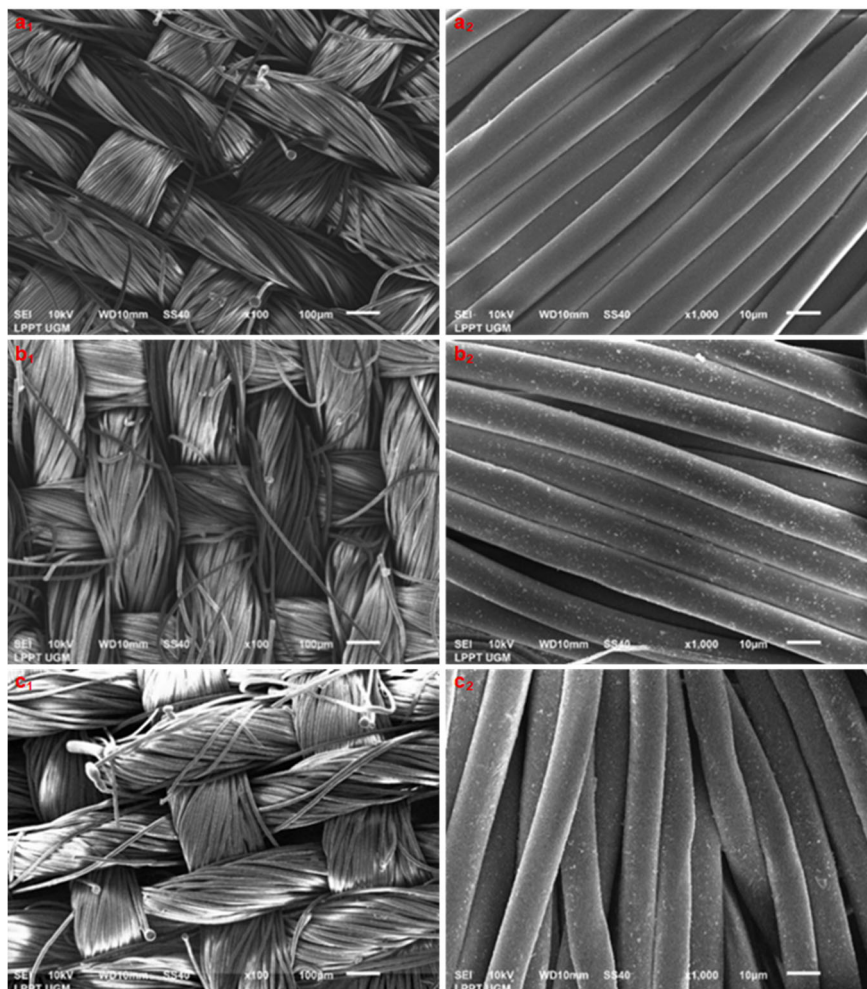


Fig 9. SEM characterization results (a) uncoated fabric, (b) PE-SO₃H/Ag-BF₃ coated fabric, (c) PE-SO₃H/Ag-H₂SO₄ coated fabric

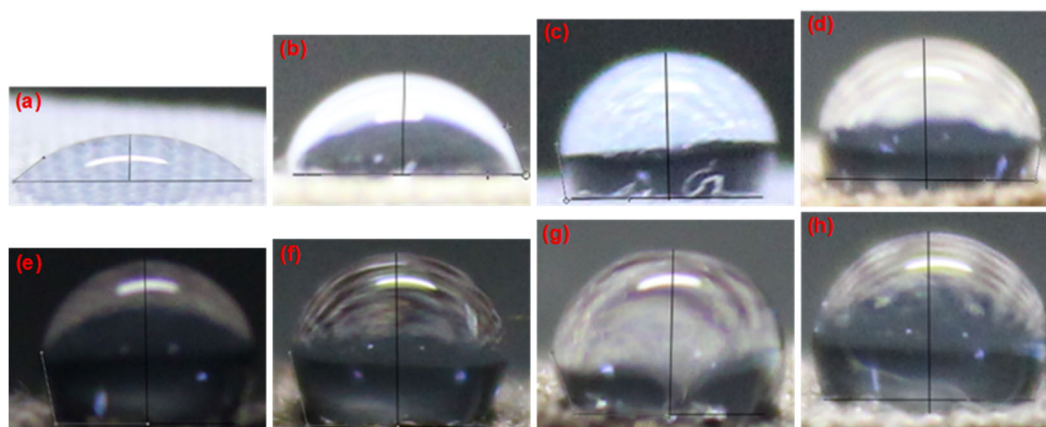


Fig 10. Contact angle results from the hydrophilicity test: (a) plain cloth, (b) Ag cloth, (c) PE-BF₃ cloth, (d) PE-H₂SO₄ cloth, (e) PE-SO₃H-BF₃ cloth (f) PE-SO₃H-H₂SO₄ cloth, (g) cloth PE-SO₃H/Ag-BF₃, (h) PE-SO₃H/Ag-H₂SO₄ fabric

Table 5. Cotton cloth contact angle value data

Sample	Contact angle (°)
Plain cloth	38.3
Cloth + Ag (1000 ppm)	69.0
Cloth + PE-BF ₃	97.2
Cloth + PE-H ₂ SO ₄	98.6
Cloth + PE-SO ₃ H-BF ₃	103.8
Cloth + PE-SO ₃ H-H ₂ SO ₄	104.5
Cloth + PE-SO ₃ H/Ag-BF ₃	109.6
Cloth + PE-SO ₃ H/Ag-H ₂ SO ₄	107.8

the impact of coating treatments on the hydrophobicity of cotton cloth, as measured by contact angle values. Pristine plain cotton cloth exhibits a relatively low contact angle of 38.3°, indicating high wettability and a hydrophilic surface. With PE modifications, especially those incorporating sulfonic acid groups and silver, significantly boost hydrophobicity. The highest contact angle of 109.6° is achieved in the sample treated with PE-SO₃H/Ag-BF₃, demonstrating superior water-repellent properties. These results prove the effectiveness of functionalized PE coatings in transforming cotton surfaces from hydrophilic to highly hydrophobic. A hydrophobic surface can minimize water binding and minimize bacterial viability. Hydrophobicity will prevent microbes from adhering to their inherent water-repellent nature. These will prevent initial bacterial adhesion and inhibit biofilm formation [19-20].

■ CONCLUSION

This study successfully demonstrates the synthesis and functionalization of polyeugenol with sulfonate groups, impregnated with Ag⁺ ions, and its applications as an antibacterial coating for cotton fabrics. The introduction of sulfonate groups improves the antibacterial activity of polyeugenol and facilitates strong interaction with silver ions. These materials synergistically improve the antibacterial activity, achieving moderate antibacterial activity confirmed by disc diffusion and turbidimetric methods. In addition, surface analysis showed uniform coating and improved hydrophobicity, which may further prevent microbial adhesion and biofilm formation. These findings highlight the promising potential of polyeugenol sulfonate/Ag⁺ as a sustainable and effective antibacterial coating for cotton fabrics, particularly in medical

applications where sterility and resistance to microbial contamination are essential.

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■ CONFLICT OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

■ AUTHOR CONTRIBUTIONS

Conceptualization, methodology, validation, resources, Muhammad Cholid Djunaidi and Khabibi; investigation, Nesti Dwi Maharani and Purbowatiningrum Ria Sarjono; writing—original draft preparation, Dandy Andhika Fatriaji; review, and editing, Nesti Dwi Maharani and Khabibi; supervision, Muhammad Cholid Djunaidi. All authors have read and agreed to the published version of the manuscript.

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