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Carboxymethyl cellulose films derived from pineapple waste: Fabrication and properties

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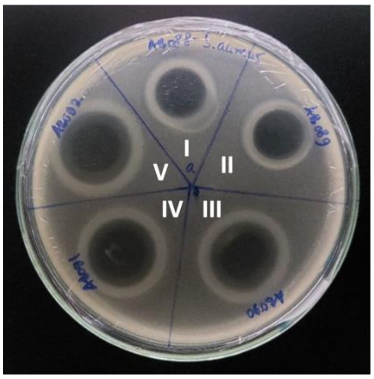

Highlights:

- Bioplastic films were developed using carboxymethyl cellulose (CMC) derived from bacterial cellulose synthesized through pineapple waste fermentation.
- Increasing glycerol concentrations improved film flexibility (up to 40.7×) but reduced tensile strength (by 89.7%), with smoother morphology and reduced crystallinity.
- The films demonstrated enhanced antibacterial activity against *E. coli* and *S. aureus*, highlighting their suitability for sustainable packaging applications.

Abstract

Plastic waste poses a significant environmental challenge due to its non-biodegradable nature, emphasizing the need for sustainable alternatives like bioplastics from natural resources. This study develops and characterizes bioplastic films made from carboxymethyl cellulose (CMC) derived from bacterial cellulose synthesized using pineapple biowaste. Pineapple waste underwent fermentation to produce bacterial cellulose, which was chemically modified into CMC. Films were fabricated using CMC solutions with varying glycerol concentrations (0.5%, 1.0%, 1.5%, and 2.5% v/v). Characterization techniques, including SEM, XRD, FTIR, TGA, mechanical testing, and antibacterial assays, revealed that increasing glycerol concentrations smoothed the film's cross-sectional morphology, reduced crystallinity, and altered functional groups (e.g., new peaks at 870 cm^{-1} and 935 cm^{-1} attributed to C–H deformation). TGA indicated a four-stage thermal degradation pattern, with mass loss increasing from 77.2% to 88.4% at 2.5% glycerol, reflecting enhanced plasticization. Mechanical testing showed that the highest glycerol concentration increased film flexibility by 40.7 times while reducing tensile strength by 89.7%. Antibacterial activity against *E. coli* and *S. aureus* also improved with glycerol content. These results demonstrate the potential of CMC-based bioplastic films as sustainable packaging materials, offering customizable properties and promoting the value-added use of agricultural waste.

Keywords: Bioplastics; Carboxymethyl cellulose; Film; Glycerol; Pineapple biowaste

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1. Introduction

In Indonesia, the potential for research and development of plastic packaging technology that can decompose in nature is very large because Indonesia is ranked 2nd in the world in biodiversity [1], but only a small amount has been utilized and has the potential to be developed into biopolymers as raw materials for bio packaging. Meanwhile, in 2022, Indonesia produced around 2.78 million tons of pineapple and become the fourth-largest pineapple-producing country in the world. Generally, 55% of pineapple fruit cannot be utilized resulting in a large amount of waste [2]. This pineapple waste can be used as a potential source of cellulose through a fermentation process [3]. One of the most common and commercial cellulose derivatives is carboxymethyl cellulose (CMC). Utilizing cellulose from alternative sources originating from pineapple peel waste is an effort to reduce forest deforestation.

CMC has attracted interest as a viable food packaging material due to its ability to produce films with desirable features such as barrier properties, tensile strength, thermal stability, and transparency [4]. However, the disadvantages of CMC, such as brittleness, need the implementation of numerous ways to improve the properties of CMC films. CMC films are delicate and weak, despite the fact that mechanical qualities are vital in determining the durability and strength of packing films [5]. In this context, chemical and physical treatments, nanoscale fillers, and polymer blends have all been extensively researched to improve CMC film characteristics [6]. Several researchers increase the flexibility of CMC film using plasticizers such as glycerol, sorbitol [7], crosslink agents (Ba^{2+}, Ca^{2+}, Zn^{2+} ions [8], and citric acid [9]), blending with other polymers like pullulan [10] and polyvinyl acetate [11] and also nanomaterials like zink oxide and graphene oxide [12]. However, plasticizer is the most suitable method to enhance the flexibility of CMC because plasticizers like glycerol are widely available, cost-effective, efficient, and practical. The process is simple, requiring only the mixing of the plasticizer into the CMC solution without the need for special conditions like high temperatures or specific pH adjustments.

According to Rachtanapun [13], CMC is a cellulose derivative used in a variety of sectors, including cosmetics, pharmaceuticals, detergents, textiles, food, and ceramics. CMC synthesis begins with cellulose extraction and purification, two key stages that have a significant impact on the quality of the resulting CMC [14]. CMC is formed by chemically modifying cellulose and adding carboxymethyl groups ($-CH_2-COOH$) to its molecular chain. Several studies reported that CMC is derived from biowaste such as almond shells and stem [15], sugarcane bagasse [16], coconut fiber [17], cotton waste [18], disposal paper [19], [20], seaweed waste [21], nata de coco [13], cassava peel [22], and empty fruit bunch [23]. Cellulose sources and also the route of CMC synthesis determine the yield and degree of substitution in CMC synthesis [24]. With the higher production of pineapple in Indonesia, it is potential to use their biowaste as a source of CMC products for maintaining a sustainable environment. The purpose of this study is to synthesize and characterize CMC film derived from pineapple biowaste with various glycerol concentrations. CMC films are characterized using SEM, XRD, FTIR, TGA, tensile testing, and antibacterial activity tests.

2. Materials and Methods

2.1. Materials

The materials used were bacterial cellulose (BC) from pineapple peel waste obtained from Blitar regency plantation (Indonesia), isopropanol (Emsure, Germany), monochloroacetic acid (Loba Chemie, India), glacial acetic acid (Emsure, Germany), methanol (Loba Chemie, India), NaOH (Loba Chemie, India), glycerol (purity 85%, Emsure, German) and distilled water.

2.2. Bacterial Cellulose Synthesis

BC was produced following procedures in a previously published study [25]. BC was produced using a medium containing a litre of pineapple peel extract, 5.0 g of ammonia acid, and 100 g of glucose, at pH 4.5. *Acetobacter xylinum* about 100 mL was added into the medium then the fermentation process was maintained at 25 °C to 30 °C. After ten days, BC floated at the upper medium and was collected and rinsed with distilled water until neutral. BC was alkalized NaOH 5% for 4 h at 70 °C. BC was crushed and homogenized using a high-pressure homogenizer machine at 150 MPa with 5 cycles.

2.3. Carboxymethyl Cellulose Film Synthesis

The production of CMC is carried out through two processes, namely alkalization and carboxymethylation. In the alkalization process, 10 grams of dry BC is added to 100 mL of isopropanol while stirring on a hotplate stirrer for 30 min. Then, 20 mL of 15% NaOH solution was added drop by drop over 1 hour while continuously stirring. After the alkalization process is complete, the carboxymethylation process is carried out by adding 7 grams of monochloroacetic acid for 3 hours at 60 °C. Then, the results of the carboxymethylation process are cooled and neutralized with glacial acetic acid until the pH reaches 7 at room temperature. CMC is filtered and soaked in 100 mL of methanol for 24 hours. CMC was filtered and dried in a dehydrator (Krisbow, Indonesia) at 60 °C. The dried CMC is then ground and stored in a closed container. The synthesis of CMC film involved dissolving 10 grams of dry CMC in 200 mL of distilled water under constant stirring for 15 minutes. Glycerol in variations of 0.5, 1.0, 1.5, and 2.5 %v/v was added in each container, and the solution is mixed until homogeneous. A CMC solution was cast onto a mold (20 cm × 15 cm) and then placed in an oven for 24 hours at 60 °C to obtain a dry CMC film. The obtained CMC film is stored in a desiccator.

2.4. Characterization of CMC Film

Determination of the degree of substitution of CMC follows the formula in the published paper [20]. The surface morphology of the film was observed using a scanning electron microscope (SEM) (Inspect S50, FEI, Japan) at a voltage of 25.0 kV. Before SEM analysis, film samples were coated with a gold layer to a thickness of 10 nm using a coater (Emitech, SC7.620, UK). The crystallinity of the CMC film was determined using X-ray diffraction (XRD) (Panalytical-Expert Pro equipment, USA). XRD analysis was conducted on samples sized 20 x 20 mm^2 with diffraction angle from 5° to 90° using a CuKα at λ of 1.542Å, 30.0 mA and 40.0 kV. The crystalline index (CI) was calculated using Eq. (1) [26]:

$$CI = \frac{I_{[200]} - I_{[am]}}{I_{[200]}} x100\% \tag{1}$$

Where: $I_{[200]}$ was the peak intensity at 2θ of about 22.0°. $I_{[am]}$ was the lowest intensity of 2θ at about 18°.

The Fourier-transform infrared spectroscopy (FTIR) investigation was carried out using an infrared equipment. The film was dried in an oven at 105 °C for 3 hours before being pulverised into a powdered sample. 1.0 mg of KBr powder was combined with CMC film powder and then compressed to produce a solid pellet. FTIR scanning analysis was conducted in wavenumber from 400 to 4000 cm^{-1} at a rate of 2.0 cm^{-1}.

Thermal degradation of CMC film was conducted in a simultaneous thermal analyzer (STA 6000. Perkin Elmer, USA) at 25 - 800 °C with a temperature rate of 10 °C /min. The mechanical properties of CMC film were evaluated using a tensile strength tester (TechnoLab. Instrument, Indonesia) with a load maximal of 50N and a pulling rate of 3.5 mm/min. The sample dimension is 10 mm in width and 65 mm in length with four replications for each film group.

2.5. Statistical Analysis

Statistical analysis was conducted using OriginPro 9.0 software (OriginLab Corp., US) to calculate the average and standard deviation values, one-way ANOVA, and Tukey test.

3. Result and Discussion

3.1. Film Fabrication

Conversion of pineapple biowaste in this conversion is 500 g biowaste results in 24 g of bacterial cellulose and after the carboxymethylation process, it can covert to 30 g of CMC (6% of biowaste weight). Synthesis of CMC results in CMC powder with a degree of substitution of 1.07. This degree of substitution is higher than CMC derived from coconut fibers (0.65) [17], bagasse (0.69) [27], Cotton gin (0.87) [18], but lower than CMC from rice hull (1.3) [28], and Almond shells (1.72) [29]. The degree of substitution (DS) in CMC (carboxymethyl cellulose) synthesis refers to the average number of hydroxyl groups on the cellulose molecule that have been replaced by carboxymethyl groups ($-CH_2COOH$) during the chemical reaction with chloroacetic acid (or its sodium salt). It is an important parameter that influences the properties and functionality of CMC that determine specific application.

CMC film without the addition of glycerol has a clear appearance, as shown in **Figure 1a**. Adding glycerol till 1.0%v/v, the film color is similar relatively to the control (**Figure 1b** and **Figure 1c**) but after adding glycerol up to a concentration of 2.5%, the resulting CMC film shows a reduction in clarity and takes on a more yellowish color, as seen in **Figure 1d** and **Figure 1e**. Improper reactions cause different results. It is thought that a higher content of glycerol causes partial miscible of glycerol. Biphasic structure caused by partial miscible also reported in starch caused by rich glycerol [30], [31].

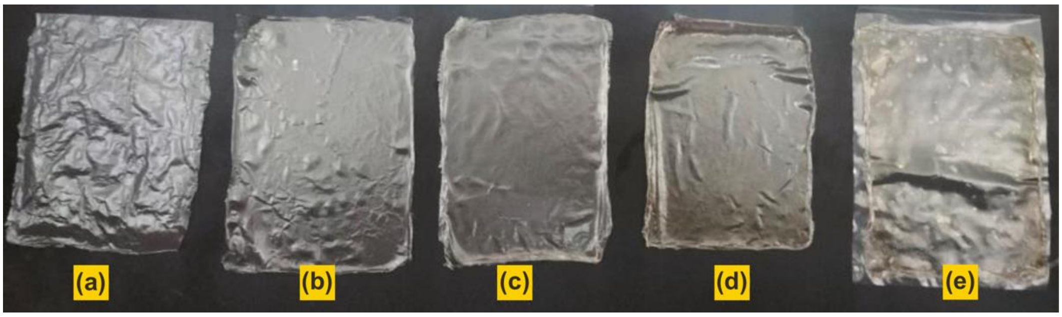

Figure 1. Product of CMC film: (a) CMC-control; (b) CMC-glycerol 0.5%v/v; (c) CMC-glycerol 1.0%v/v; (d) CMC-glycerol 1.5%v/v; (e) CMC-glycerol 2.5%v/v

Agricultural biowaste has been recycled and transformed into energy, and composite materials for many applications in lightweight parts which provide both environmental and economic benefits [32]. The cost of production is one of the parameters productions of bioplastic. The production of synthetic plastics uses more energy input than bioplastics. The cost of manufacturing petrochemical-based plastics, which is 78–88 MJ/kg of resin is much higher than that used for the fossil fuel-based energy required to produce bioplastics, 44 MJ/kg of bioresin [33], as well as, the cost of material production of synthetic fiber is higher than natural fibers [34]. However, while the initial cost of synthetic plastic might be higher, the price of synthetic plastic products is lower than bioplastic with the average price between USD 3.35 and USD 4.69 per kg, which is slightly costlier than that of other conventional polymers [35] due to the mature industry, large-scale production, and low raw material costs, even though the initial production cost is higher. In contrast, bioplastic products are more expensive despite being environmentally friendly because of developing technology and smaller production scales.

3.2. Surface Morphology

The detail of the film surface was investigated by SEM, with the results displayed in **Figure 2**. Without a plasticiser, CMC bioplastics were stiff, brittle, and fragile with a wavy texture (**Figure 2a**). This could be ascribed to strong intramolecular hydrogen interactions that limited macromolecular chains' mobility, resulting in a fragile, stiff surface that led the films to break apart. The film CMC control has a smooth surface, but after adding 0.5% glycerol, the surface seems rough, with many wave ripples (**Figure 2b**) and many strips are shown in cross section surface. Increasing the glycerol concentration to 1.0%v/v. results in the roughest surface with less wave ripple, but high peak wave ripple (**Figure 2c**) and a number of strips is reduced in cross section surface. When glycerol is increased by 1.5%, the wave ripple becomes smoother (**Figure 2c**), and when glycerol is increased by 2.5%, the film surface becomes smoother and tends to produce beach marks and cross section surface also become smooth.

Glycerol, a plasticiser, is known to contain low-molecular-weight molecules [36]. Glycerol interacts with CMC molecules, increasing the mobility of CMC chains and reducing polymer chain bonding [37], modifying the surface shape of bioplastic film. This change is also seen in the cross-sectional area of film. Glycerol has a comparable effect on smoothing the surface of a bioplastic film formed of palm starch [38]. A higher glycerol content smoothens the cross-sectional area and reduces brittle features.

3.3. Functional Group Analysis

Functional group analysis was conducted using FTIR. The IR spectrum indicates the functional bond of the chemical structure of molecules in the sample. The IR absorption bands in the spectrum of 4,000-400 cm^{-1}. The results of FTIR test are as shown in **Figure 3**. FTIR spectra of CMC film control was denoted by black curve, and CMC film with glycerol content of 0.5%, 1.0%, 1.5%, and 2.5% represented by the red, blue, brown, and dark green curves, respectively.

Some obvious changes were detected after adding CMC with glycerol. The change curve from the broad peak observed in 4,000–3,600 cm^{-1} corresponds to OH-stretching vibration, significant in determining hydrogen bonding [39], [40]. Adding glycerol of 2.5% to the CMC film causes reduce in the transmittance at the wavenumber of 2360 cm^{-1} band, wave number of 2360 cm^{-1} corresponds to the aliphatic C–H group [41]. C-H stretching vibration in the 2700–3200 cm^{-1} range from the cellulose [42]. Notably, the transmittance of O-H stretching appears comparable, suggesting that adding glycerol does not alter the hydrophilicity of the film. The transmittance of wavenumber 1665 cm^{-1} is reduced, which belongs to the C=O, representing the metal-oxygen vibration band [43]. The wavenumber of 1520-1400 cm^{-1} indicates the bending CH_2, C-H, and C-O groups in cellulose [44].

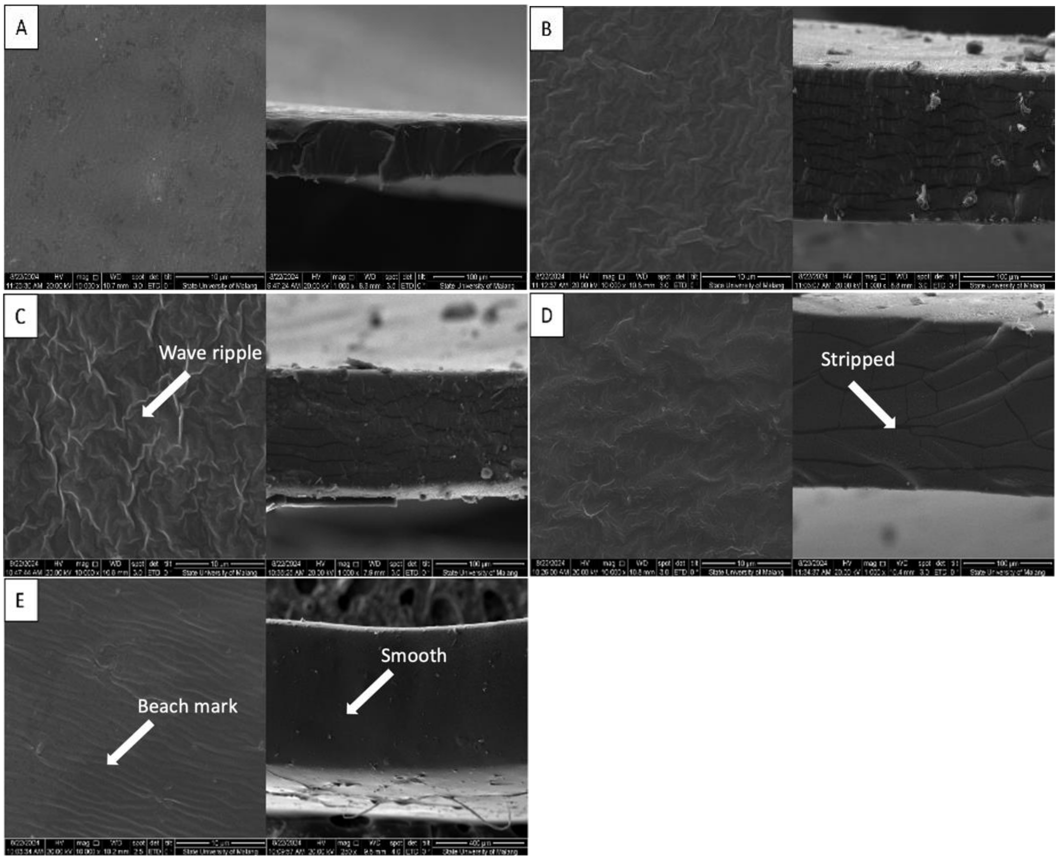

Figure 2. Morphology of CMC film of: (a) CMC-Control; CMC with glycerol of (b) 0.5%v/v; (c) 1%v/v; (d) 1.5%v/v; (e) 2.5%v/v (left side: surface film; right side: cross-section film)

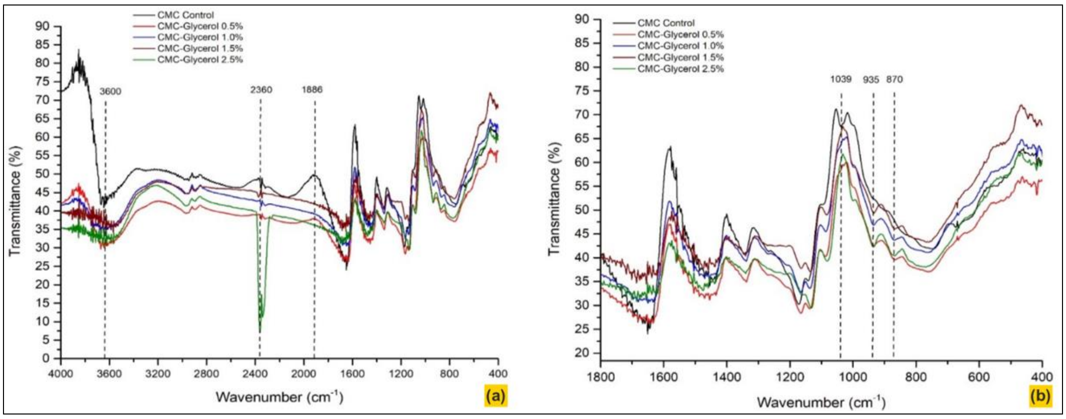

Figure 3. FTIR analysis of (a) In wavenumber 4000-400/cm; (b) In wavenumber 1800-400/cm

Wavenumber ranges from 950 to 1250 cm^{-1} indicating ether bonds in the CMC compound [45]. The wave number 1040 cm^{-1} was characteristic of the C-O stretching on polysaccharide skeleton in CMC [46]. The intensity of this peak is less obvious with the addition of glycerol. New peaks rose at a wavenumber of 870 and 935 cm^{-1}. These peaks are in the 834–950 cm^{-1} range and are typically assigned to C–H deformation vibrations [47]. These peaks might also indicate interactions between CMC and glycerol, as glycerol can influence the conformation and flexibility of CMC polymer chains, potentially affecting the vibrational modes in this range.

3.4. Crystalline Structure Analysis

Figure 4 shows the diffractogram of CMC film, with a large peak at approximately 22°. It highlighted the polymer's amorphous behaviour. After adding glycerol, it reverted to an amorphous state, with the peak shifting at a diffraction angle of around 22°. Peaks in the 2θ range (10°-75°) showed minor change, but intensity reduced with increasing glycerol concentration. The addition of glycerol to the CMC film was rather weak, most likely due to its growth on the CMC film's surface, which made it difficult to gather diffraction peak data during the XRD test. However, utilising a larger glycerol content resulted in lower crystallinity because glycerol alters the hydrogen-bonding interactions in the film [48].

Segal equation is used to calculate the CI of cellulose-based material [26]. This CI serves as an indicator of the crystalline structure organized in the film. According to Utari, bacterial cellulose membrane had CI of about 89% [49]. The analysis results showed that CI of CMC film is 52.5% (**Table 1**) or lower than CI of bacterial cellulose as based CMC film materials. It indicates that disrupt crystalline structure of cellulose after carboxymethylation of cellulose. These phenomena are also shown in carboxylated cellulose nanocrystals with CI of about 61.1% [50]. CMC with the addition of glycerol generally had a lower CI value than the control CMC. The adding glycerol of 0.5%, 1.0%, 1.5%, and 2.5% into CMC reduced the CI film with values of 46.6 %, 56.2 %, 46.1%, and 46.0%, respectively. These findings collectively demonstrate that the glycerol content contributed to an overall decrease in the crystalline nature of CMC film, which could have significant implications for the final properties of film.

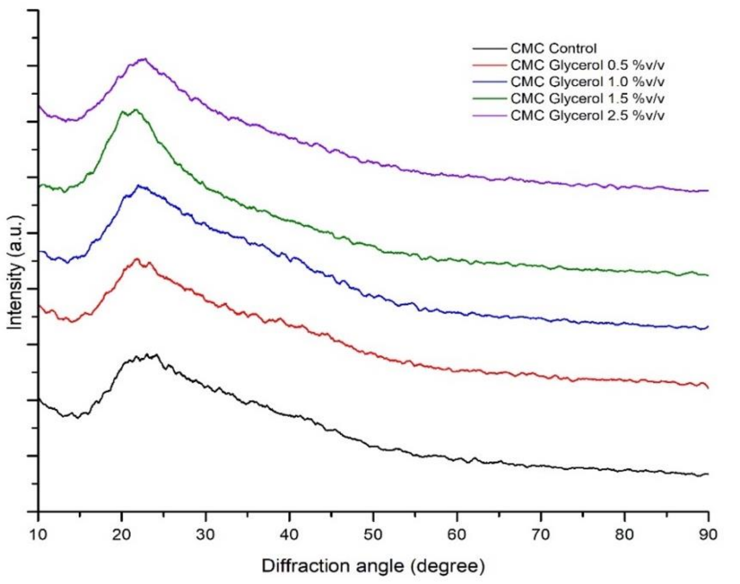

Figure 4. XRD graph of CMC film with various glycerol content

Table 1. Crystallinity of CMC film

Sample	Diffraction angle (°)	I_{002} (a.u.)	I_{am} (a.u.)	CI (%)
CMC-Control	22.8	139	66	52.5
CMC-Glycerol 0.5 %v/v	22.4	147	80	45.6
CMC-Glycerol 1 %v/v	22.1	169	91	46.2
CMC-Glycerol 1.5 %v/v	22.0	191	103	46.1
CMC-Glycerol 2.5 %v/v	22.7	150	81	46.0

3.5. Thermal Decomposition Analysis

The stability of CMC film at elevated temperatures was analysed using a thermogravimetric analyzer (TGA). The thermogravimetric (TG) and differential thermogravimetric are indicate the decomposition of CMC film control and adding glycerol of 0.5, 1.0, 1.5, 2.5 %v/v after subjected to heat are shown in **Figure 5a** to **Figure 5e**, respectively.

The film indicates a multi-stage decomposition process attributed to Stage I, II, III, and IV (**Figure 5a**). Stage I was observed at a temperature below 233 °C (Stage I), which represents the evaporation of water content and devolatilization of the film at evaporation peak at 72.4 °C and massa loss in this stage about 21.7%. Increasing glycerol content in the film increases the mass loss of volatile matter to 27.9% after adding glycerol 2.5% (**Table 2**). It confirms that adding glycerol into CMC makes the film bind more easily to the water. It is confirmed by FTIR analysis results that indicate a decrease in the percentage of transmittance of a hydroxyl group (**Figure 2a**). Glycerol has hydrophilic properties that easily bind to water than CMC [21].

Stage II represents the depolymerization process of the CMC film with a mass loss of 30% for the control sample and an increase of up to 50% for CMC film with glycerol 2.5%. The peak of decomposition occurs at 287.2 °C with a mass rate decomposition of 10.11%/min. After adding glycerol as a plasticizer into CMC, the peak decomposition temperature and total solid decomposition parameter of TG and DTG changed. After adding glycerol of 0.5, 1.0, 1.5, and 2.5 %v/v, peak decomposition temperature becomes 268.2, 267.7, 270.9, and 289.6 °C with mass rate decomposition also reduced to 7.12, 7.16, 5.69, and 6.05 %/min, respectively. Interestingly, after adding glycerol, there are 2 peaks of decomposition in stage II (**Figure 4a** to **Figure 4e**) with maximum peak change along with increasing the concentration of glycerol. The lower (269 °C), corresponds to the initial decomposition of glycerol that is incorporated into the CMC matrix. It is probably related to the partial miscibility of glycerol in the CMC, consistent with the color change of the films (**Figure 1**) and the deformations observed SEM (**Figure 2**). Pure glycerol has a burn-out temperature of 238–276 °C [51], lower than the decomposition temperature of CMC ranging from 250 to 360 °C [22]. Glycerol, acting as a plasticiser, reduces the initial decomposition temperature by breaking the connections between the CMC polymer chains. As a result, certain components of the mixture (mostly glycerol or CMC segments that interact closely with glycerol) begin to decompose at a lower temperature than pure CMC. The second peak corresponds to the primary disintegration of the CMC matrix. Due to the presence of partial immiscible of glycerol, the decomposition temperature of CMC may be slightly elevated. Glycerol can form hydrogen bonds with CMC, requiring higher temperatures for breakdown.

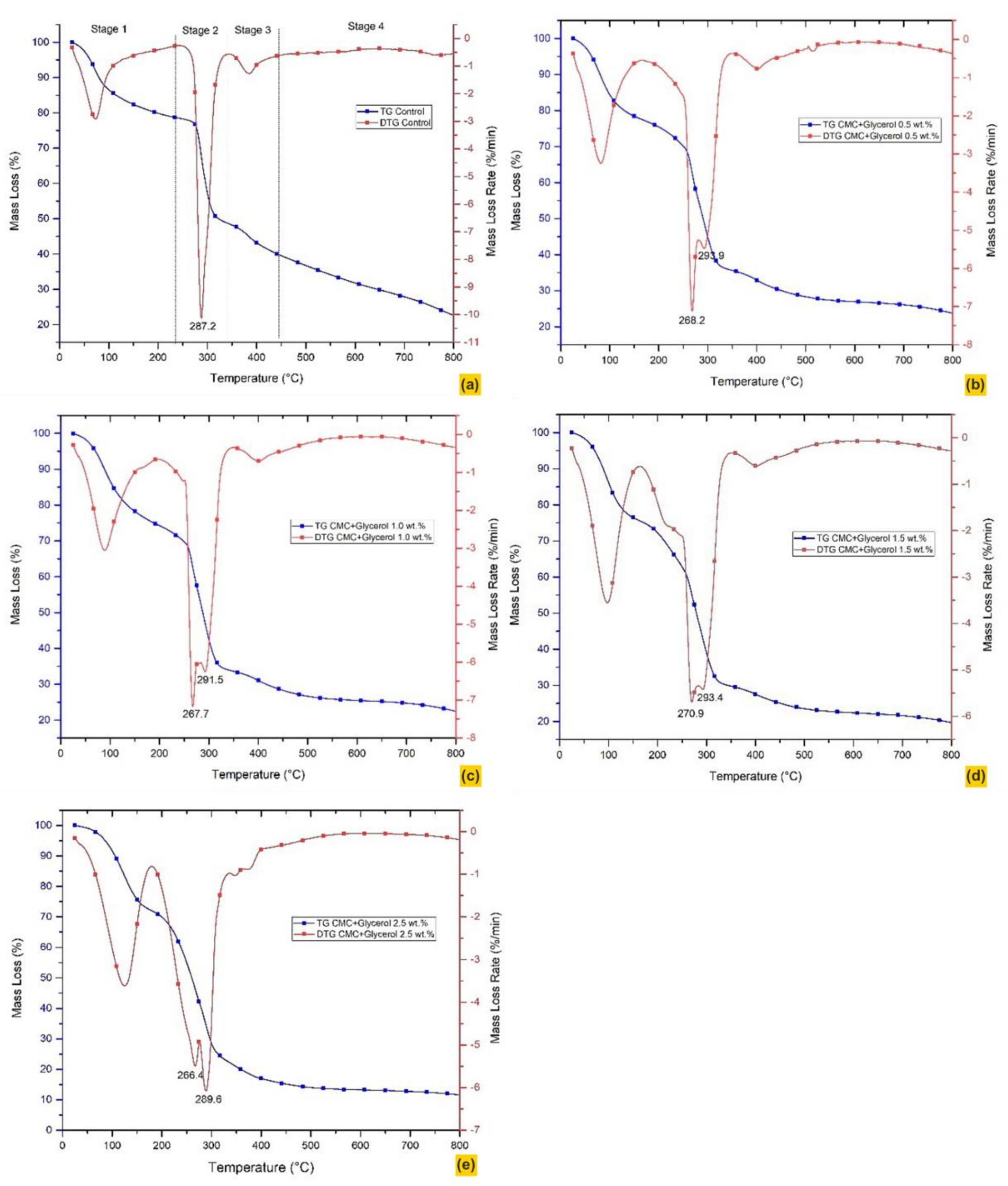

Figure 5. Thermal decomposition analysis of CMC film of: (a) Control; (b) 0.5 %v/v; (c) 1.0 %v/v; (d) 1.5 %v/v; (e) CMC with 2.5 %v/v

Stage III indicates further solid decomposition in the 340 – 450 °C. After that, the film tends to become ash (Stage IV). The total mass loss of CMC control film is about 77.2% (**Table 2**). After adding glycerol with the concentrations of 0.5, 1.0, 1.5, and 2.5 %v/v, into CMC, total mass loss is become 76.2, 77.5, 80.2, and 88.4%, respectively. This result indicates that adding glycerol into CMC film results in increasing stability temperature of CMC film.

Increasing the amount of glycerol enhanced the film's hydrophilicity due to increased hydroxyl groups, as confirmed by **Figure 3a**. Glycerol retains moisture within the CMC film [52], contributing to greater mass loss during the initial heating stages as water evaporates. These effects collectively increase the overall thermal mass loss observed in glycerol-plasticized CMC films, as confirmed in thermogravimetric analyses showing distinct peaks for water evaporation and organic decomposition. Furthermore, glycerol decreases strong intermolecular hydrogen bonding in CMC [21], weakening the polymer structure and facilitating its thermal degradation over a broader temperature range. An increasing rate of thermal degradation is also identified in starch film with increasing glycerol concentration [53] as well as starch content [54]. In addition, the conversion of cellulose to CMC results in a higher mass loss compared to cellulose from hemp fiber as much as 74.5% [55].

Table 2. Mass rate and mass loss in thermogravimetric analysis

Treatment CMC film	Peak of volatile released (°C)	Mass rate max. volatile released (%/min)	Mass volatile released (%)	Peak of decomposition (°C)	Mass rate max. decompose (%/min)	Total mass loss (%)
Control	72.4	2.92	21.7	287.2	10.11	77.2
Gly 0.5	79.9	3.24	22.4	268.2	7.12	76.2
Gly 1.0	89.2	3.06	24.2	267.7	7.16	77.5
Gly 1.5	96.5	5.55	24.8	270.9	5.69	80.2
Gly 2.5	125.5	3.61	27.9	289.6	6.05	88.4

3.6. Mechanical Properties

The tensile test result for CMC film in comparison to other published results is shown in **Table 3**. The use of glycerol affects the mechanical properties of CMC film where increasing glycerol concentration causes decreasing in the tensile strength and elongation at break of the film. The tensile strength of the control sample is about 49.72±3.69 MPa. This strength is higher after comparing to CMC film derived from palm bunch, CMC from culture standards, and commercials that have a tensile strength of 5.68 ± 1.67 MPa [16], and 38.13 MPa [56], and 58±5 MPa [57], respectively. This strengtht is higher than bioplastic from blending cassava and corn starchas much as 0.04 MPa [58]. After adding glycerol of 0.5, 1.0, 1.5, and 2.5 %v/v, tensile strength reduced to 31.20±4.84, 7.69±1.95, 6.39±0.90, and 5.12±1.66 MPa or reduced 18.5%, 84,5%, 87.1% and 98.7%, respectively. But, the strain or elongation at break of CMC film increases from 2.63±0.54% (control) to 12.66±2.11%, 35.68±5.78%, 42.47±2.83%, and 109.80±6.60%, or increase about 381%, 1257%, 1519%, and 4070%, after adding glycerol of 0.5, 1.0, 1.5, and 2.5 %v/v, respectively. Shiva et al suggest that immiscible polymer with other components causes intrinsic incompatibility and weak interfacial adhesion resulting in decreasing tensile strength [21], [59]. Several researchers report decreasing tensile strength after adding glycerol into the CMC matrix derived from various sources [16], [56], [57].

Statistical analysis using one-way ANOVA indicates that film thickness is not significantly different with P_{value} of 0.104 (**Table 3**). The addition of glycerol of 0.5, 1.0, 1.5, and 2.5 %v/v into CMC film have significantly different tensile strengths, elongation at break and elastic modulus with a confidence level of 95% (P_{value} = 0.00). Further analysis using Tukey methods indicates that no different tensile strength results after treatment using treatment among 1.0, 1.5, and 2.5 %v/v. It means after a concentration of 1.0%v/v, glycerol results in a relatively similar tensile strength of CMC film. The addition of glycerol with a concentration of 0.5 %v/v has no difference value compared to the control as well as the concentration of glycerol of 1.0 %v/v compared to 1.5 %v/v. The addition of glycerol in all concentrations results in no significant difference in elastic modulus except in the control.

Table 3. Properties of CMC film derived from pineapple biowaste

Sample	Thickness (µm)	Tensile Strength (MPa)	Elongation at Break (%)	Elastic Modulus (MPa)	Ref.
Control	108±13^a	47.04±10.8^a	2.63±1.0^a	1848.03±265.1^a	This study
CMC+gly. 0.5%	89±46^a	27.57±8.0^b	12.67±33.2^a	215.64±76.6^b	This study
CMC+gly. 1.0%	164±56^a	7.69±3.4^c	35.68±10.0^b	20.95±4.3^b	This study
CMC+gly. 1.5%	135±33^a	6.40±1.6^c	42.47±4.9^b	14.93±2.3^b	This study
CMC+gly. 2.5%	129±13^a	5.12±2.9^c	109.80±11.4^c	4.81±3.1^b	This study
Palm bunch + gly.0.3%	213 ± 052	5.68 ± 1.67	56.18 ± 26.17	-	[16]
Bagasse + gly.0.3%	111 ± 11	13.94 ± 1.1	79.08 ± 12.59	-	[16]
CMC Commercial	87 ± 5	58±5	28±3	6.74±0.06	[57]
CMC+gly 0.5% cake extract 40%	140 ± 10	3±2	156±15	0.29±0.07	[57]
BC+ gly 0.6%	-	6.70	-	52.5	[56]

Note: values in column followed by the same letter (a, b, and c) are not significantly different (n=3, $p > 0.05$)

Glycerol, as a tiny, flexible molecule, disrupts the hydrogen connections between the CMC polymer chains. This rupture decreases the stiffness of the polymer matrix, allowing the chains to move more freely past each other under stress. Glycerol is a versatile chemical that can take on many different shapes [60]. Glycerol breaks the hydrogen bonds between CMC polymer chains. When glycerol is introduced to CMC, it interacts with the polymer chains and breaks the hydrogen bonds between neighbouring CMC polymer molecules [21]. This disruption bonding weakens the polymer chains, making them less stiff and allowing them to move more easily. This disruption adds to the CMC film's greater flexibility and elongation under stress. Several researchers have found a similar phenomenon, in which an increase of plasticisers reduces the mechanical characteristics of bioplastic film. According to Fransiska et al., glycerol reduced the tensile strength of seaweed film while increasing its flexibility [61]. A comparable report was published for collagen fiber/CMC film [62], chitosan/CMC film [48], starch/glycerol/oleic acid film [63], and cellulose film [64].

In the present results, the increasing glycerol content in the CMC matrix has shown an increase in both the flexibility and thermal stability of CMC film. As the glycerol content increased, the flexibility of the CMC system improved due to the reduction in intermolecular forces between the polymer chains [21], which allowed greater molecular mobility. CMC film exhibited enhanced flexibility, likely due to glycerol's plasticizing effect. Interestingly, despite the reduction in intermolecular forces, the decomposition temperature (**Table 2**) of the material also increased with higher glycerol concentrations (2.5%v/v). This can be attributed to the partial immiscibility of glycerol with CMC, where the immiscible phases likely formed regions that absorbed heat and delayed the thermal degradation process. Consequently, the system showed a shift in the decomposition temperature to higher values, suggesting that glycerol content not only increased the material's flexibility but also contributed to its enhanced thermal stability [65], likely by absorbing heat and preventing premature breakdown of the CMC matrix. This dual effect highlights the complex interactions between glycerol and CMC, where glycerol acts as both a plasticizer and a thermal stabilizer, enhancing the material's performance across different properties.

3.7. Antibacterial Activity

The prepared samples' antibacterial activity against E. coli and S. aureus was evaluated, with clear zone diameters measured to assess inhibition. The results of the antibacterial test are depicted in **Figure 6**. A positive control (Chloramphenicol) demonstrated the highest antibacterial efficacy, serving as a positive control (**Table 4**).

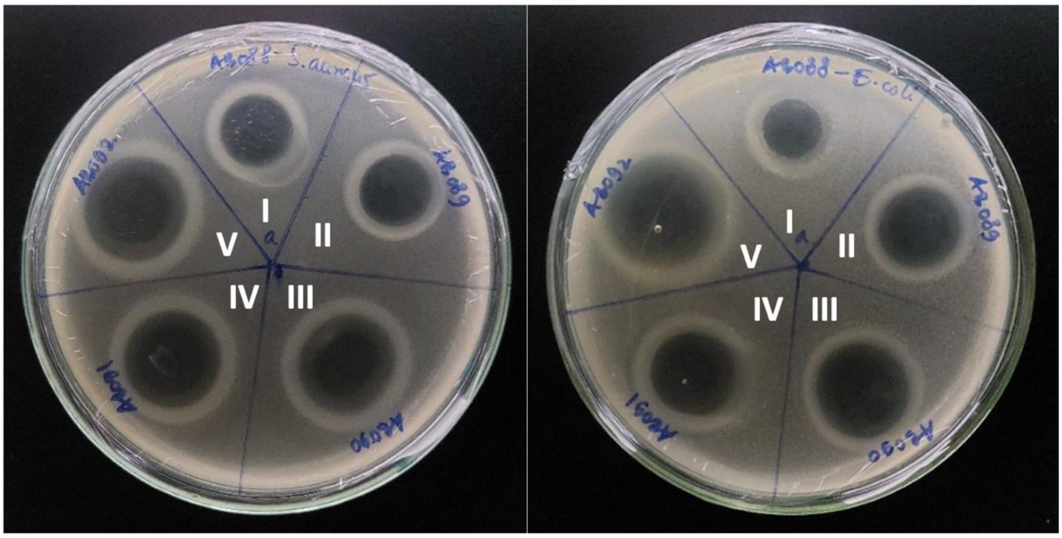

Figure 6. Antibacterial activity of CMC film against *E.coli* (left side) and *S.aureus* (right side): I, II, III, IV, and V: control, glycerols content of 0.5, 1.0, 1.5, and 2.5%v/v, respectively

Table 4. Measurement of antibacterial activity of CMC film with various glycerol content

Samples	Clear zone diameter (mm)	
	E.coli	***S.aureus***
Control positive (Chloramphenicol)	22.32	21.85
CMC Control	14.6 ± 2.25^{a}	12.1 ± 0.75^{a}
CMC Glycerol 0.5%	11.5 ± 3.67^{a}	11.7 ± 3.23^{a}
CMC Glycerol 1%	12.2 ± 5.07^{a}	$15.9 \pm 0.78^{a/b}$
CMC Glycerol 1.5%	$19.3 \pm 3.47^{a/b}$	$19.9 \pm 1.60^{b/c}$
CMC Glycerol 2.5%	25.4 ± 2.07^{b}	21.8 ± 1.87^{c}

Note: values in column followed by the same letter (a,b, and c) are not significantly different (n=3, $p > 0.05$)

One-way ANOVA analysis (**Table 4**) indicates that the addition of glycerols significantly differs in antibacterial activity against E.coli with P_{value} of 0.003 and S.aureus with P_{value} of 1.74 10^{-4}. The Tukey analysis reveals that the addition of glycerol significantly influences the antibacterial activity of the CMC films, particularly at higher concentrations (2.5%v/v) showing the best performance that approaches the efficacy of Chloramphenicol. The antibacterial activity against *E. coli* and *S. aureus* follows a similar trend, but there are slight differences in significance levels across the groups. *E. coli* appears slightly more sensitive to the treatments, as reflected in the larger clear zone diameters for corresponding groups. Lower glycerol concentrations do not significantly differ from the CMC control.

Antibacterial properties of glycerols are reported in bacterial cellulose matrix [66], [67]. Although glycerol is not an active antibacterial agent, in CMC films, it can enhance antibacterial activity through osmotic effects that cause dehydration in bacteria. Adding CMC film containing glycerol has more hydroxyl groups (see **Figure 3a**) which can interact with water by hydrogen bonds, more water molecules can enter the membranes, and changing the osmotic conditions or moisture levels of bacteria environment. Bacteria are sensitive to the changes in their environment [68] and reducing moisture levels significantly decreased bacterial growth [69].

Abundant glycerols in film result in a hypertonic environment where the concentration of solutes outside the bacterial cell is higher than inside. This difference drives water out of the bacterial cell through osmosis, leading to dehydration and cytoplasmic shrinkage with enlargement of regions of the periplasm, a process known as plasmolysis [70]. Limiting water availability inhibits enzyme activity and nutrient transport [71], which are essential for metabolic processes and cellular growth, while also compromising the structural integrity of the cell membrane. Over time, dehydration causes irreversible damage to cellular components [72], resulting in bacterial death.

4. Conclusion

We were able to successfully characterise the utilisation of pineapple biowaste for the production of bacterial cellulose for CMC film with a variety of glycerol concentrations. After adding glycerol up to 1.0% volume/volume, the film demonstrates that the opacity and morphology of the film are rough. However, after adding a larger percentage of glycerol, the surface morphology demonstrates increased smoothness. After the addition of glycerol, the crystallinity of the film decreases as a consequence of the interaction between glycerol and CMC, which is demonstrated by changes in the FTIR curve. A higher temperature makes the film more stable, and the addition of glycerol at a ratio of 2.5% by volume. The mass loss of the film is increased to 88.4% as a result of decomposition. The higher content of glycerol increases antibacterial activity against *E.coli* and *S.aureus,* significantly. It shows a difference in film flexibility significantly with the highest elongation of 40.7 times obtained by glycerol content of 2.5%v/v, however, the tensile strength of the film is decreased by 98.7% and elastic modulus decreases with reducing glycerol content. In the future, this film could be further explored as a potential candidate for use in packaging applications with high flexibility, antibacterial activity, and heat resistance of bioplastic films of CMC derived from pineapple biowaste.

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Authors' Declaration

Authors' contributions and responsibilities - **HS:** Funding Acquisition, Project Administration, Writing – original draft, and Writing - Review and Editing. **DS**: Writing – review & editing, Visualization, Resources, Methodology, and Investigation. **AF**: Validation, Resources, Methodology, and Investigation; **UY:** Writing – review & Editing, Validation, Methodology, and Investigation. **JSB**: Writing – review & editing, Visualization, and Methodology. **FN**: Visualization, Resources, Methodology, and Investigation. **K**: Visualization, Resources, Methodology, and Investigation. and **UAU**: Visualization, Resources, Methodology, and Investigation.

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Availability of data and materials - All data is available from the authors.

Competing interests - 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.

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