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THE EFFECT OF CARBOXYMETHYL CELLULOSE AND GLYCEROL ADDITION ON THE BIODEGRADABILITY OF CASSAVA STARCH-BASED BIOFILMS

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ABSTRACT

The growing concern over plastic pollution has accelerated the development of biodegradable packaging materials derived from natural polymers. Cassava starch has been widely studied as a potential raw material because of its abundance and biodegradability. However, its properties are often modified with additives such as carboxymethyl cellulose (CMC) and glycerol to improve film performance, which may influence its biodegradation. This study aimed to investigate the effects of different concentrations of CMC and glycerol on the biodegradability of cassava starch-based biofilms. Biofilms were prepared using three levels of cassava starch (5, 10, and 15 g), three concentrations of CMC (1, 2, and 3%), and three levels of glycerol (6, 9, and 12 mL). The biodegradability test was performed using the soil burial method for seven days, with weight loss as the primary parameter. The results showed that all biofilm samples achieved more than 60% degradation within seven days, thus meeting the Indonesian National Standard (SNI) for biodegradability. At low starch concentrations (5 g), the addition of higher CMC and glycerol levels reduced biodegradability due to matrix compaction and reduced porosity. At 10 g starch, biodegradability was more stable, with several formulations reaching 100% degradation by day seven. The highest starch concentration (15 g) yielded the best results, with nearly all formulations reaching complete biodegradation, supported by sufficient substrate availability and synergistic interactions between CMC and glycerol. Overall, the findings indicate that starch concentration plays a dominant role, whereas excessive CMC and glycerol can slow degradation by limiting microbial accessibility.

Keywords: Biodegradability, Biofilm, Carboxymethyl Cellulose, Cassava Starch, Glycerol

INTRODUCTION

Conventional plastics pose a major environmental challenge because of their resistance to natural degradation, leading to long-term accumulation and pollution (Jiang et al., 2020). Indonesia ranks among the world's largest contributors to plastic waste, with significant amounts entering both terrestrial and marine ecosystems. This underscores the urgent need for sustainable alternatives to conventional plastic films. One potential solution lies in biodegradable plastics, particularly biodegradable films, which can be decomposed by microorganisms into environmentally benign products (George et al., 2021).

Among the various forms of biodegradable plastics, biodegradable films have attracted particular attention owing to their wide application in packaging, especially as replacements for conventional plastic films. Cassava starch is considered a highly promising biodegradable film due to its abundance, affordability, and high starch content, which can reach up to 90% (Cheng et al., 2021). Moreover, Indonesia, as one of the largest cassava-producing countries, has significant potential to utilize this resource for the development of biopolymers. Nevertheless, starch-based films generally suffer from limitations, such as low mechanical strength and poor flexibility, necessitating the incorporation of reinforcing and plasticizing agents.

In starch-based biodegradable films, the incorporation of a plasticizer is essential to overcome the inherent brittleness caused by strong intermolecular hydrogen bonding within the polymer matrix (Hidayat et al., 2023). Glycerol is one of the most widely used plasticizers in biodegradable film systems because of its low molecular weight, high compatibility with hydrophilic biopolymers, and multiple hydroxyl groups that enable effective interaction with starch and cellulose derivatives. By partially replacing polymer–polymer hydrogen bonds with polymer–glycerol interactions, glycerol increases chain mobility, resulting in improved flexibility, elongation, and film integrity (Giubertoni et al., 2023). However, glycerol is hygroscopic, and excessive concentrations can increase water uptake and reduce the water resistance of the films (Affanti et al., 2024). Therefore, optimizing the glycerol content is critical for achieving a balance between mechanical flexibility and barrier performance in starch–CMC biodegradable films.

Carboxymethyl cellulose (CMC) has been widely applied as a reinforcing filler to improve the mechanical and barrier properties of starch-based films (Cui et al., 2021), whereas glycerol is commonly used as a plasticizer to enhance flexibility and reduce brittleness (Fahrullah and Ervandi, 2022). However, the inclusion of these additives may also influence the biodegradability of the films, which is a critical parameter that determines their environmental compatibility.

This study aimed to analyze the effects of varying concentrations of cassava starch, CMC, and glycerol on the biodegradability of cassava starch-based biofilms. The results were evaluated with reference to the Japanese Industrial Standards (JIS) and Indonesian National Standards (SNI), contributing to the development of biofilm formulations that balance functional performance with environmental sustainability.

MATERIALS AND METHODS

Materials

Cassava starch was extracted locally and used as the primary raw material for the study. Carboxymethyl cellulose (CMC) and glycerol were used as additives to modify the film properties. Distilled water was used to prepare the film-forming solutions, and garden soil was used as the medium for the soil burial biodegradability test.

Experimental Design

A factorial experimental design was applied with three levels of cassava starch concentration (5, 10, and 15 g), three levels of CMC concentration (1, 2, and 3% w/w based on starch), and three levels of glycerol volume (6, 9, and 12 mL). This resulted in 27 unique treatment combinations being generated.

Biofilm Preparation

Biofilm-forming solutions were prepared by dissolving a specified amount of cassava starch in distilled water and heating the mixture to 75 °C until gelatinization. Subsequently, CMC and glycerol were added in accordance with the experimental design and stirred until homogeneous. The mixtures were then cast onto molds and dried in an oven at 60°C for 24 h to obtain thin biofilms. This method was used by Brion-Espinoza et al. (2021) with minor modifications to the original method.

Biodegradability Test

A biodegradation test was performed using soil microorganisms to assist in the degradation process, which is known as the soil burial test (Natalia et al., 2019). The biodegradability of the biofilm samples was evaluated using the soil-burial method. Film specimens measuring 3 × 3 cm were buried at a depth of 10 cm in garden soil, with soil moisture maintained at approximately 50–60% of water-holding capacity and temperature controlled at 25–30 °C to ensure consistent microbial activity. The samples were retrieved daily over a seven-day period, carefully cleaned of soil residues, dried, and weighed to monitor their weight losses over time. The percentage of biodegradation was determined by comparing the initial dry weight of each sample before burial with the dry weight after retrieval, expressing the loss as a percentage. This method provides a quantitative measure of the biofilm degradation rate in a natural soil environment.

Biodegradability was measured using the following equation:

$$\text{Biodegradability (\%)} = \frac{\text{Initial weight } - \text{Final weight}}{\text{Final weight}} \times 100$$

This formula was used to calculate the biodegradability of the samples based on weight changes during the degradation process (Acharjee et al., 2023).

RESULTS AND DISCUSSION

This biodegradability test was conducted to determine the ability of soil microorganisms to degrade plastic films. The test was carried out using The Soil Burial Test method, in which the samples were buried in soil and their weights were observed before and after burial for seven days (Kuswytasari et al., 2019). All cassava starch–based biofilm samples fulfilled the Indonesian National Standard (SNI) requirement of >60% degradation within seven days, with several formulations achieving complete degradation in less than a week.

Starch concentration was the dominant factor influencing biodegradability: higher starch levels (15 g) provided an abundant substrate for microorganisms, resulting in the fastest and most uniform degradation, whereas low starch levels (5 g) made the films more sensitive to the effects of CMC and glycerol. At 5 g starch, excessive CMC and glycerol reduced porosity and slowed degradation, whereas at 10 g starch, biodegradability became more stable, and several formulations achieved full degradation by day 7 (Fig 1.).

At 15 g starch, nearly all formulations reached 100% degradation, with the optimal formula of 15 g starch, 1% CMC, and 6 mL glycerol showing complete degradation by day five. Overall, starch content primarily controlled degradation, whereas appropriate levels of CMC and glycerol enhanced flexibility, porosity, and microbial accessibility. These findings highlight the importance of balancing starch concentration with additive ratios to produce biofilms that are both mechanically stable and environmentally degradable, underscoring their potential for sustainable food-packaging applications.

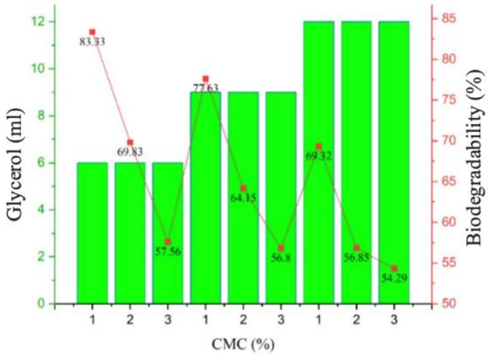

Figure 1. Graph of the effect of the CMC and glycerol combination on the biodegradability test rate with 5 g cassava starch concentration.

In the cassava starch-based films containing 5 g of starch, the addition of CMC and glycerol significantly influenced biodegradability. CMC reinforced the film structure, which tended to slow down the degradation process, particularly at higher concentrations. Meanwhile, glycerol, as a plasticizer, increased the flexibility of the film; however, excessive glycerol led to a more homogeneous matrix with reduced porosity, making it more difficult for microorganisms to access the starch substrate.

At low starch concentrations, the main components, amylose and amylopectin, were relatively accessible; therefore, the effects of CMC and glycerol became more dominant in determining the biodegradation rate. The optimal combination of 1% CMC and 6 mL glycerol showed the highest biodegradability, as the film matrix remained sufficiently porous and flexible to facilitate the microbial activity. Conversely, formulations with higher concentrations, such as 3% CMC and 12 mL of glycerol, exhibited lower biodegradability (down to 83.09%) owing to a denser and more complex polymer network. Overall, the combination of CMC and glycerol at low starch levels enhanced film porosity, solubility, and flexibility, collectively accelerating microbial access and improving the biodegradation rate.

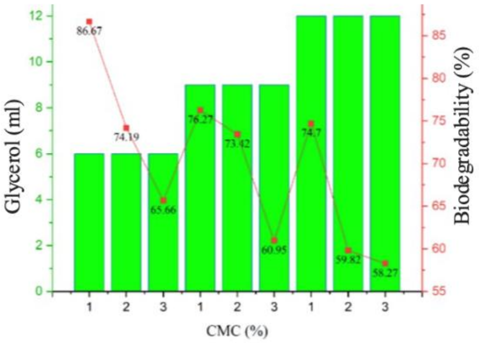

Figure 2. Graph of the Effect of CMC and Glycerol Combination on Biodegradability Rate with 10 g Cassava Starch Concentration

Figure 2 shows that for 10 g of cassava starch, the combination of CMC and glycerol affects biodegradability in a manner similar to that of 5 g of starch, but with notable differences owing to the increased film matrix density. The highest biodegradability reached 100% on day 7 for certain combinations, such as 1% CMC with 6 mL of glycerol. However, at higher concentrations (e.g., 3% CMC and 12 mL glycerol), biodegradability increased to 87.07%, not due to a higher starch content, but due to the enhanced accessibility of starch chains to microbial and enzymatic attacks (Kuswytasari et al., 2019).

Although CMC forms a denser polymer network through hydrogen bonding with starch, the high glycerol content increases matrix plasticization, water uptake, and free volume, facilitating microbial penetration and accelerating the biodegradation process. The difference arises from the matrix structure, porosity, and plasticization rather than the starch quantity (Ramakrishnan et al., 2024). CMC acts as a thickener and matrix-strengthening agent, forming a denser polymer network through hydrogen bonding with starch, which limits microbial access and slows degradation.

At high CMC concentrations, the matrix becomes rigid, reducing biodegradability (Fig. 3). Similarly, glycerol enhances film flexibility but reduces porosity, creating a more homogeneous matrix that is harder to penetrate by microorganisms. Compared to 5 g starch, the 10 g starch films had a denser and more organized matrix, which balanced mechanical strength and flexibility. This allows for higher and more stable biodegradability, especially with optimal combinations of CMC and glycerol (e.g., 1% CMC with 6–9 mL glycerol), as starch molecules are more available and accessible to microorganisms. However, high additive concentrations can negatively affect biodegradability by increasing matrix density and limiting microbial access.

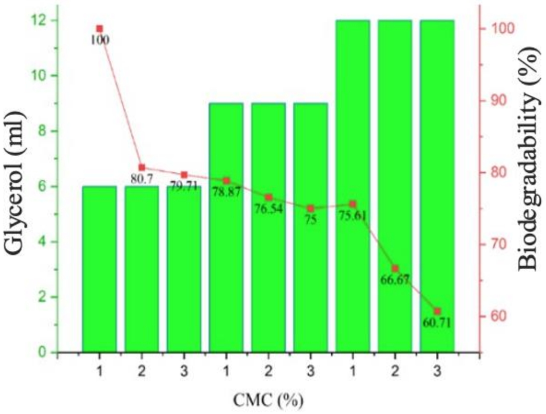

Figure 3. Effect of CMC and glycerol on biodegradability of 15 g cassava starch films

The graph in Figure 3 shows that films containing 15 g of cassava starch exhibited very high biodegradability, with most formulations reaching 100% degradation by day 7. Higher starch concentrations provide abundant substrates in the form of amylose and amylopectin, facilitating microbial activity and accelerating degradation (Munfarida and Hidayat, 2023). Despite the denser matrix compared to 10 g starch films, glycerol contributed to flexibility and sufficient porosity, allowing microorganisms to access the starch effectively. CMC strengthens the polymer network while maintaining hydrophilicity, improving water absorption, and softening the film structure, which enhances microbial access. Glycerol, a plasticizer, increases matrix flexibility, creates intermolecular spaces, and attracts moisture, further supporting the biodegradation process (Dewi et al., 2021). The synergistic combination of CMC and glycerol is suggested to enhance matrix flexibility and permeability, which can facilitate water diffusion and microbial access even at high starch concentrations (Munfarida and Hidayat, 2023), thereby promoting efficient biodegradation. Compared to the 5 g and 10 g starch films, the 15 g starch films showed the highest and most stable biodegradability owing to the greater substrate availability and optimal matrix properties. All cassava starch-based films met the Indonesian National Standard (SNI) for biodegradability, requiring at least 60% degradation within a week, with some formulations achieving complete degradation in less than seven days. The 15 g starch film with 6 mL of glycerol performed best, reaching 100% degradation in less than a week, demonstrating its eco-friendly potential and suitability for food packaging applications.

The three graphs (Fig 1–3) collectively illustrate the influence of cassava starch concentration on the biodegradability of the films in combination with CMC and glycerol. For films containing 5 g starch, biodegradability was moderately high but variable. Low starch concentrations created a loosely organized matrix, rendering the films highly sensitive to the effects of CMC and glycerol. At this level, CMC reinforced the film structure, whereas glycerol increased the flexibility. However, excessive concentrations of either additive reduced biodegradability by creating denser or overly homogeneous matrices, thereby limiting microbial access to the starch substrate. The optimal combination (1% CMC and 6 mL glycerol) resulted in the highest biodegradability, suggesting enhanced matrix permeability and chain mobility, which facilitated microbial activity. In films with 10 g starch, biodegradability became more stable and was generally higher than that in the 5 g starch films. The increased starch content provides a larger substrate pool of amylose and amylopectin, allowing microorganisms to degrade the polymer network more effectively (Tapia-Blácido et al., 2022). Although high concentrations of CMC and glycerol slightly reduced biodegradability by stiffening the matrix or reducing porosity, many formulations, including 1% CMC with 6–9 mL of glycerol, reached complete degradation (100%) by day 7. The denser but more organized matrix at this starch concentration balanced structural strength with flexibility, supporting more consistent microbial access across the film. The biodegradability of the films containing 15 g starch was the highest and most uniform among all concentrations. Almost all the formulations reached 100% degradation by day 7. The abundant starch substrate enhances microbial activity, whereas the synergistic effects of CMC and glycerol maintain a flexible and porous matrix despite the higher polymer density (Bergel et al., 2020). CMC reinforced the network and improved water absorption, facilitating matrix softening, while glycerol further increased flexibility and hygroscopicity, promoting microbial penetration. Notably, the formulation with 15 g starch and 6 mL glycerol exhibited the fastest and most complete degradation, highlighting the optimal balance between the polymer concentration and additive effects.

Overall, the comparison of the three starch levels demonstrated that increasing the starch concentration enhanced biodegradability by providing more substrate for microorganisms and stabilizing the matrix. At low starch concentrations, additive effects dominate, occasionally hindering biodegradability. At higher starch levels, the matrix was robust yet accessible, allowing CMC and glycerol to synergistically improve the film properties without compromising degradation. These results confirm that the proper adjustment of starch concentration and additive ratios is crucial for achieving films that are both mechanically stable and environmentally degradable.

CONCLUSION

All cassava starch–based biofilms met the SNI requirement of >60% degradation within seven days, with several formulations achieving complete degradation in less than a week. Starch concentration was the key determinant of biodegradability, as higher starch levels (15 g) supplied more substrate for microbial activity, resulting in faster and more uniform degradation, whereas lower starch levels (5 g) made the films highly influenced by CMC and glycerol content. Conversely, at low starch concentrations, the effect becomes more pronounced because of the inherently less dense polymer matrix. A low CMC content (1%) provides sufficient network stabilization without excessively limiting microbial access. Excessive additives tend to create denser matrices that limit microbial access, whereas optimal combinations improve porosity, flexibility, and water absorption to support degradation. At 10 g starch, biodegradability became more stable, and several formulations reached 100% by day 7, whereas at 15 g starch, nearly all formulations achieved full degradation, with the best result (15 g starch, 1% CMC, and 6 mL glycerol) degrading completely by day 5. These findings confirm that the proper adjustment of starch concentration with CMC and glycerol is essential to balance mechanical stability and environmental degradability, making cassava starch biofilms a promising material for sustainable packaging applications.

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