

# Optimization of Cassava Starch–CMC–Glycerol Formulations for Enhanced Water Resistance of Biofilms

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**Abstract.** Biodegradable films from natural polymers offer a sustainable alternative to synthetic plastics, with water resistance being a critical property. This study examined the effects of cassava starch (5 g, 10 g, 15 g), carboxymethyl cellulose (CMC: 1%, 2%, 3%), and glycerol (6 mL, 9 mL, 12 mL) on biofilm water resistance, producing 27 formulations. Water resistance was measured through immersion tests by assessing weight changes due to absorption and swelling. Results showed that starch concentration strongly influenced matrix formation, where higher starch content (15 g) produced denser and more stable films with greater resistance. CMC reduced water resistance because of its hydrophilic nature, while glycerol acted as a plasticizer with dual effects: moderate amounts improved flexibility and minimized cracks, but higher levels increased water uptake. Among all formulations, only the film with 15 g starch, 1% CMC, and 6 mL glycerol met the Indonesian National Standard (SNI), achieving 103.3% water resistance. These findings highlight the importance of optimizing starch, CMC, and glycerol composition to produce biofilms with strong barrier properties for biodegradable packaging applications.

## 1 Introduction

### 1.1 Background

The increasing consumption of conventional plastics has become a global environmental challenge, particularly in Indonesia, one of the largest contributors of plastic waste to the oceans. Conventional plastics are non-biodegradable, requiring hundreds of years to decompose, and their incineration releases harmful emissions that contribute to environmental pollution and human health risks [1], [2]. The widespread production and disposal of fossil-based plastics have led to substantial accumulation of plastic waste in terrestrial and aquatic ecosystems, raising global concerns about ecosystem degradation and waste management challenges [3]. Plastic pollution adversely affects wildlife and ecosystems while increasing greenhouse gas emissions and dependence on finite fossil resources, further underscoring the unsustainability of current plastic use patterns. Consequently, there is an urgent need for environmentally friendly materials that can reduce reliance on petrochemical-derived plastics and mitigate long-term ecological impacts. In this context, biodegradable plastics (bioplastics) derived from renewable natural polymers have been proposed as a promising and more sustainable alternative, offering potential benefits such as decreased carbon footprint and enhanced degradability under appropriate conditions [4].

Cassava starch is widely available in Indonesia and contains about 17% amylose and 83% amylopectin, making it suitable for biodegradable film production (Putri et al., 2021). Its abundance in Balikpapan and surrounding areas makes cassava a strategic local raw material. However, starch-based films are hydrophilic and have low mechanical strength, so additives

need to be added. CMC can exhibit gelation properties, which means that it can act as a thickener, stabilizer, binder and form a fine texture with the aim of improving the mechanical characteristics of the biodegradable film formed [5] and is able to bind with water so as to minimize shrinkage of the plastic or increase the ability to bind water. Among various additives investigated to improve the performance of bioplastics, carboxymethyl cellulose (CMC) is frequently selected due to its excellent compatibility with biopolymer matrices, ability to enhance mechanical and barrier properties, and its renewable, biodegradable, and non-toxic nature, making it particularly suitable for sustainable packaging applications [6].

Cellulose and its derivatives play a crucial role in biodegradable films, functioning as polymer matrices, reinforcing agents, and performance-enhancing additives. Owing to their renewable origin, biodegradability, strong hydrogen-bonding capability, and tunable physicochemical properties [7]. Cellulose-based materials significantly improve the mechanical strength but the resulting film tends to be stiff [8]. For this reason, a plasticizer is added which functions to provide plastic properties to the biodegradable film, reduce brittleness due to its rigid nature, increase flexibility and durability of the film made, especially when stored at relatively low temperatures. The plasticizer used in this research is glycerol. Glycerol was chosen as an effective plasticizer to use because it has a good ability to reduce internal hydrogen bonds in intermolecular bonds. Biopolymers such as cellulose, starch, and proteins contain many hydroxyl ( $-OH$ ) groups that form strong intermolecular hydrogen bonds. While these bonds provide strength, an excessive hydrogen-bonding network restricts polymer chain mobility, resulting in stiff and fragile films that easily crack under stress [9]. Plasticizer has low volatility, when added it will provide flexibility and elasticity of the polymer chain in the resulting biodegradable film. According to [10] glycerol increases the flexibility and tensile strength of starch-based films compared to sorbitol or polyethylene glycol (PEG).

Based on the research gap, this research aims to optimize the formula of cassava starch-based biodegradable film with the addition of CMC as filler and glycerol as plasticizer. The main focus of the research is to improve the mechanical strength, flexibility and water resistance of the biofilm so that it can be used as an environmentally friendly and economical alternative to food packaging. Thus, this research not only answers environmental challenges due to plastic waste, but also encourages innovation of local products based on renewable resources, while supporting the economic sustainability of the community in the agricultural sector.

## **2 Materials and Methods**

### **2.1 Materials**

The tools used in this research include grater, filter cloth, blender, oven, hot plate magnetic stirrer, analytical balance, 100 mesh sieve, thermometer, stopwatch, and petri dish. The materials used are cassava tubers (as starch source), carboxymethyl cellulose (CMC), glycerol, distilled water and silica gel (moisture control).

### **2.2 Starch Extraction**

Starch extract was obtained by peeling, washing, shredding cassava tubers, and filtering with water (1:2 b/v). The suspension was precipitated for 8 hours, the starch precipitate was re-washed, dried at 60°C oven for 72 hours, then sieved with 100 mesh. Starch extraction method obtained from [11], [12] by modification.

### **2.3 Biofilm Preparation**

Solution casting method was employed. Cassava starch (5, 10, or 15 g) was dissolved in 150

mL distilled water with stirring at 380 rpm. Glycerol (6, 9, or 12 mL) and CMC (1, 2, or 3%) were added sequentially. The solution was heated at 75 °C for 10 min, poured into trays, and dried in an oven at 60°C for 24 hours. Biofilm casting method obtained from [13] by modification.

## 2.4 Water Resistance

This test is conducted to determine the occurrence of bonds in the polymer as well as the level or regularity of bonds in the polymer which is determined through the percentage of polymer weight gain after swelling. The process of diffusing solvent molecules into the polymer will produce a bulging gel. The nature of bioplastic resistance to water is determined by the swelling test, which is the percentage of film swelling in the presence of water to test the resistance of biodegradable plastic to water [14]. Measurement of water resistance is done using the following equation:

$$\text{Water (\%)} = \frac{W-W_0}{W_0} \times 100\% \tag{1}$$

This equation is used to calculate the sample's resistance to water based on the weight changes that occur during the degradation process [15].

Description:

- A = Water absorption (%)
- W = Test weight after immersion (gr)
- W0 = Initial test weight

Then, the percentage of water absorbed is calculated again in the following calculation to get the percent water resistance.

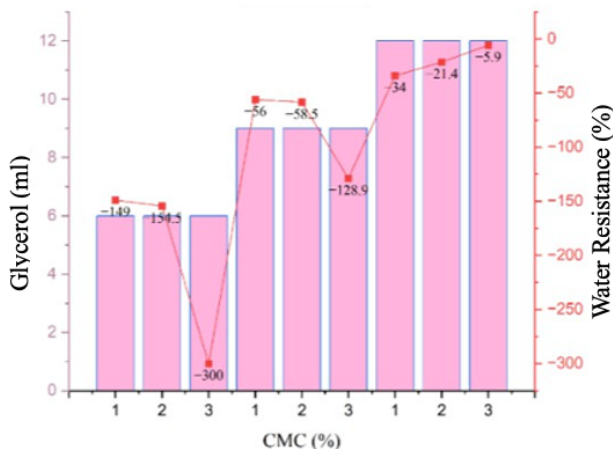
$$\text{Water Resistance (\%)} = 100\% - \text{Water Absorbed} \tag{2}$$

The test was conducted by cutting the film sample into dimensions of 2 cm × 2 cm, followed by recording its initial weight (W0). The sample was then immersed in a Petri dish containing 30 mL of distilled water for 3 minutes. After immersion, the sample was carefully removed, surface-dried using tissue paper, and subsequently weighed to obtain the final weight (W). The percentage of water absorbed was then calculated based on the difference between W0 and W.

## 3 Results and Discussion

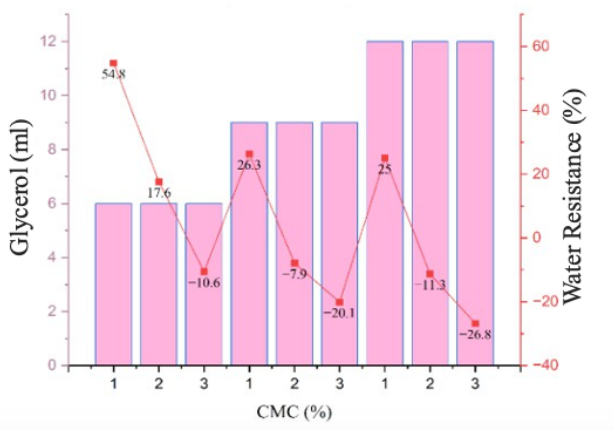
Water resistance test to determine the occurrence of bonds in polymers and the level or regularity of bonds in polymers determined through the percentage of polymer weight gain after swelling. The nature of bioplastic resistance to water is determined by the swelling test, which is the percentage of film swelling in the presence of water to test the resistance of biodegradable plastic to water.

Water resistance is calculated by weighing the weight of the bioplastic when swelling occurs due to the diffusion of water into the bioplastic. Water resistance is needed in the manufacture of bioplastics to be applied industrially and commercially as an alternative to synthetic plastics. The results of research data on the effect of the combination of cassava starch formula, CMC, and glycerol on water resistance are presented in the following graph.



**Figure1.** Effect of CMC and glycerol Formula Combination on Water Resistance Test of Film with 5 gr Cassava Starch Concentration

On the 5 gr cassava starch sample, the data showed that the addition of CMC generally decreased the water resistance value of the film. This is due to the hydrophilic (water-loving) nature of CMC which increases water absorption in the film matrix. As a water-soluble polymer, CMC forms a polymer network that facilitates the penetration of water molecules into the film structure. However, in samples 7-9 (12 mL glycerol), the water resistance value actually increased compared to other combinations. This happens because the high amount of glycerol creates a more homogeneous and flexible matrix, thus reducing micro-cracks that are usually the pathway for water to enter. The interaction between glycerol and CMC at this concentration results in a more structured polymer network, although it remains hydrophilic [14]. Which means, glycerol is a plasticizer capable of reducing stiffness and increasing the flexibility of the biofilm. At high concentrations (12 mL), glycerol helps the CMC and starch molecules to arrange themselves in a more organized network. This means that the interactions between the biofilm constituent molecules (CMC, starch, and glycerol) become stronger and more coordinated, making the biofilm matrix more stable [8]. Since glycerol increases flexibility, micro-cracks that usually form in rigid biofilms can be minimized. These micro-cracks are usually a way for water to enter into the biofilm structure.

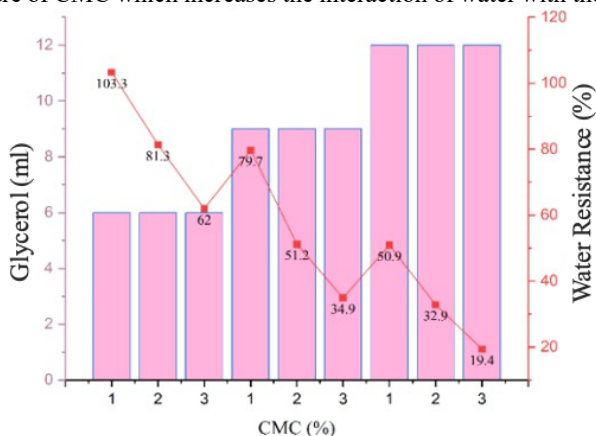


**Figure 2.** Effect of CMC and Glycerol Formula Combination on Water Resistance test of Film with 10 gr Cassava Starch Concentration

On the 10 gr cassava starch sample, CMC was seen to be very constant in reducing the water resistance value of the film, in contrast to what was seen in the 5 gr starch. This is because, at higher starch concentrations such as 10 gr the film matrix formed is denser and more organized. Increasing the concentration of CMC in starch-based films with higher amounts of starch can increase water penetration as the denser network tends to absorb water more easily [16].

Meanwhile, glycerol constantly decreased the water resistance in all samples at 10 gr of starch. As a plasticizer, glycerol increases the flexibility of the matrix by softening the polymer network, but at the same time, its hygroscopic properties make the film more susceptible to absorbing water from the environment [17]. At 10 gr starch, glycerol creates a more homogeneous matrix, which, while improving flexibility, also makes it easier for water to penetrate into the film structure.

The role of cassava starch in the 10 gr sample is significant in forming a denser matrix compared to 5 gr. At a concentration of 10 gr, the starch creates a more stable matrix base, but this structure is still affected by the natural hydrophilic nature of the starch itself, which makes it susceptible to interaction with water [18]. In the 15 gr cassava starch sample, CMC consistently decreased the water resistance value, just like in the 10 gr sample. This is due to the hydrophilic nature of CMC which increases the interaction of water with the film matrix.



**Figure 3.** Effect of CMC and Glycerol Formula Combination on Water Resistance Test of Film with 15 gr Cassava Starch Concentration

Glycerol also constantly lowers the water resistance of each sample due to its hygroscopic properties (ability to absorb water). As a plasticizer, glycerol softens the polymer network to increase flexibility, but at the same time makes the matrix more susceptible to absorbing water. At a starch concentration of 15 gr, glycerol amplifies this effect by creating a homogeneous film structure yet more open to water absorption. [19] noted that glycerol can weaken the water resistance of starch-based films due to its property of attracting water from the environment. The role of cassava starch in the 15 gr sample is to provide a very dense and mechanically stable matrix, with amylose molecules providing linear strength and amylopectin increasing flexibility [20]. At this concentration, starch is the main component that forms the film structure, but its hydrophilic properties also contribute significantly to the interaction with water. Starch, although hydrophilic, binds water in a more structured and preserved form.

Table 1. Samples that Meet SNI on Water Resistance Test Results

SNI (%)	Composition (Starch : CMC : Gliserol)	Water Resistance Test Results	Description
99	15 : 1 : 6	103,3	Meets the Indonesian National Standard

Based on the data obtained, the water resistance of biofilms in various combinations of cassava starch, CMC, and glycerol formulas showed significant variations. Only sample 19, with the composition of starch: CMC : glycerol (15:1:6), met the Indonesian National Standard (SNI) for water resistance, with a minimum value of 99%. This sample showed a water resistance of 103.3%, which means it was able to withstand water uptake very well. Meanwhile, the other samples, although showing some positive values, were mostly still below the standard or even showed negative values, reflecting greater water absorption compared to water loss. The water resistance of biofilms is greatly influenced by the composition of the formulation, especially the interaction between cassava starch, CMC, and glycerol. Cassava starch, as the main component of the film matrix is hydrophilic so it tends to more absorb water.

The fluctuations in the data which are increases and decreases indicated the water resistance properties of the biofilm are influenced by the complex interactions between the concentrations of cassava starch, CMC, and glycerol. This change in value depends not only on one component individually, but on how the three components work synergistically or against each other in forming the biofilm matrix. An increase in water resistance value occurs when the formulation produces a tighter and more homogeneous matrix, where fillers such as CMC fill the gaps in the film structure, and glycerol in optimal concentrations improves flexibility without damaging the structure. Conversely, a decrease in water resistance value occurs if one of the ingredients, such as excessive glycerol or CMC in too high a concentration, disturbs the balance of the matrix, so that the film becomes more porous or too soft, allowing greater water penetration.

It also reflect that water resistance is greatly influenced by the balance between the hydrophilic and hydrophobic properties of the components. For example, the close-packed nature of cassava starch at high concentrations can improve water resistance, but this effect can be compromised by the hygroscopic nature of glycerol or the hydrophilic nature of CMC if not in the right proportion. Previous research, by [21], also showed that the mechanical and physical properties of biofilms are not only affected by the amount of a component, but by the interactions between components and the method of preparation. Therefore, the main conclusion from these data is the importance of formula optimization to achieve the ideal balance between cassava starch, CMC and glycerol. With a systematic approach, biofilm formulation can produce consistent properties suitable for application as an environmentally friendly food packaging material.

## 4 Conclusion

The study demonstrated that water resistance of cassava starch-based biofilms is strongly influenced by the interactions among starch, CMC, and glycerol. CMC consistently reduced water resistance due to its hydrophilic nature, while glycerol, although acting as a plasticizer to improve flexibility, lowered resistance at higher concentrations because of its hygroscopic

properties. Increasing starch concentration from 5 gr to 15 gr improved matrix density and stability, contributing to better water resistance, though starch itself remained hydrophilic. Among all formulations, only the composition of 15 gr starch, 1% CMC, and 6 mL glycerol met the Indonesian National Standard (SNI), achieving 103.3% water resistance. These findings highlight that optimal proportions of starch, CMC, and glycerol are essential to balance flexibility, porosity, and hydrophilicity, thereby producing biofilms with reliable barrier properties. A systematic optimization of these components can lead to consistent and functional biodegradable films suitable for environmentally friendly food packaging applications.

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