

Optimization of Carboxymethylcellulose (CMC) for Environmentally Friendly TNI Rations Packaging Formulation Based on Polysaccharides

Fajar Ismail Rhomadon¹, Anggi Khairina Hanum Hasibuan^{1,*}, Tedi Kurniadi¹, Chrisna Prawiradhiva Ramadhanty¹, Putry Wanda Azzida¹

¹ Department of Chemistry, The Republic of Indonesia Defense University, Bogor, Indonesia

* Corresponding Author: anggi.hasibuan@idu.ac.id

Abstract

Conventional plastic waste is difficult to decompose and has the potential to leave traces in the TNI field of operations. This condition has encouraged the development of polysaccharide-based bioplastics as a safe, environmentally friendly, and easily degradable alternative for ration packaging. This study aims to optimize the concentration of carboxymethylcellulose (CMC) as a filler in the formulation of polysaccharide bioplastics for TNI ration packaging. The method used was a literature review on the effect of CMC addition on the physical and mechanical properties of bioplastic films. The results of the study showed that the addition of CMC generally increased tensile strength and water absorption, but also accelerated dissolution and the rate of biodegradation. The optimum concentration varied depending on the type of polymer used. Therefore, the selection of CMC concentration must be adjusted to the application objective, whether to increase mechanical strength or accelerate biodegradation, and needs to be validated through mechanical and degradation tests under real conditions. Compliance with bioplastic quality standards and the use of safe additives are highly recommended for application in TNI ration packaging.

Keywords: Bioplastics, Carboxymethylcellulose, TNI Rations

Introduction

Most Indonesians are consumptive in their use of plastic. Based on data obtained from the Indonesian Plastic Industry Association (INAPLAS) and the Central Statistics Agency (BPS), as cited by Puspita (2018) [1], Indonesia produces approximately 64 million tons of plastic waste each year, with 3.2 million tons of it being disposed of in the sea. Therefore, Indonesia is recognized as one of the largest contributors of plastic waste in the ocean, with a total of 187.2 million tons, after China [2]. This causes many negative impacts on the environment.

Plastic, commonly used for food packaging, is economical; however, it has several disadvantages. Plastic is difficult to decompose in nature, taking around 500 to 1000 years to decompose [3]. Therefore, plastic waste accumulation causes many negative impacts, such as killing marine life and reducing soil fertility [4]. When burned, plastic produces air pollution and releases toxic substances into the air that can be inhaled by humans. Plastic waste can also be a factor in flooding if it is disposed of in sewers and rivers [5].

Plastic is one of the factors that hinders the movement of the Indonesian

National Armed Forces (TNI) in the field. The TNI is required not to leave any traces so that their whereabouts are difficult for the enemy to detect. One example is the plastic packaging used for TNI rations [6]. This is inefficient and requires alternative packaging that is easily degradable, environmentally friendly, and leaves no trace. One solution to address this problem is to replace conventional plastic with biodegradable bioplastic [7].

Bioplastic is plastic that can be broken down by microorganisms, 10 to 20 times faster than conventional plastic [8]. Bioplastics can be made using biomass materials containing polysaccharides such as starch and cellulose [9]. The most commonly used polysaccharide is cellulose obtained from plants [10].

In the manufacture of bioplastics, additional materials are required, one of which is filler to increase tensile strength. Carboxy Methyl Cellulose (CMC) is a commonly used filler. The concentration of CMC added affects the physical and chemical characteristics of the resulting bioplastic. Therefore, optimization of the CMC concentration is needed to produce the best bioplastic, which can be an alternative for TNI ration packaging that is safe, environmentally friendly, and leaves no trace.

Methods

This review was conducted using the literature review method. This method involves critically examining all ideas, knowledge, and new discoveries in credible literature in order to answer the questions that are the purpose of the research paper. The research data was collected, organized, and grouped, and then compared with the applicable standards to complete the research. The Indonesian National Standard (SNI) for Easily Degradable Plastic Bags was used [33]. The search for published scientific articles was conducted using Google, Google Scholar, and ResearchGate with the keywords bioplastics, biodegradable plastics, fillers, and CMC.

Results and Discussion

Packaging materials

Food packaging materials are made from various types of compounds, depending on the type of need. Plastic is the most commonly used packaging material. Plastic has many advantages, such as being rust-resistant, strong, lightweight, and elastic [10]. This has led to an increase in the use of plastic packaging in Indonesia, causing negative impacts on human health and environmental pollution. Plastic can be classified into four groups, as shown in Table 1.

Table 1. Classification of Materials Based on Biodegradability and Biobased

| Aspect | Group 1 | Group 2 | Group 3 (conventional) | Group 4 |
|----------------------|--|--|---|--|
| Chemical Composition | <ul style="list-style-type: none"> • Polyamide (PA), • Polytrimethylene Terephthalate (PTT) | <ul style="list-style-type: none"> • Polylactic acid (PLA), • Polyhydroxyalkanoate (PHA), • Polybutylene succinate (PBS), • starch or cellulose blends | <ul style="list-style-type: none"> • Polyethylene (PE) • Polypropylene (PP) • polyethylene terephthalate (PET) | <ul style="list-style-type: none"> • polybutylene terephthalate (PBAT) • Polycaprolactone (PCL). |
| Source | Biobased: <ul style="list-style-type: none"> • ammonia (NH₃) and amines (R-NH₂) • Terephthalic acid and trimethylene glycol | Bio-based: <ul style="list-style-type: none"> plant polysaccharides (starch, cellulose, PBS) or biological polyesters (PLA, PHA) | Fossil-based: <ul style="list-style-type: none"> Terephthalic acid and ethylene glycol. Terephthalic acid is made from crude oil, while ethylene glycol is produced from petroleum ethylene. | Fossil-based: <ul style="list-style-type: none"> adipic acid, terephthalic acid, and 1,4-butanediol • Caprolactone |
| Chemical bond | <ul style="list-style-type: none"> • repeating amide bonds (-CONH-) in its molecular structure. • Covalent bonds between monomers | Ester bonds (PLA, PHA), glycosidic bonds (starch, cellulose), hydrogen bonds, carbon bonds easily broken down by microbes | Covalent bonds between carbon-hydrogen, carbon-carbon (PET, PP), | Ester bonds |
| Biodegradability | <ul style="list-style-type: none"> • Does not have chemical bonds that can be degraded by microbial enzymes • Presence of additives • Modification of the main raw material | Can be broken down by microorganisms through the breaking of ester or glycosidic bonds | Cannot be broken down by microorganisms because the covalent bonds are very stable | Can be broken down by microorganisms that have esterase enzymes |
| Toxicity | May contain hazardous additives such as heavy metals | Generally non-toxic, more environmentally friendly | May contain harmful additives such as phthalates or heavy metals | Low toxicity, more environmentally friendly |

(Source: SNI: 7188-7: 2022, Pandey et al., 2020, Indriani et al., 2023, Pasaribu et al., 2021 [27])

This table compares plastic classifications based on their biodegradability and primary material sources. Conventional plastic materials commonly used for food packaging can pose environmental problems because they are generally not easily broken down naturally and are long-lasting. Emissions are produced by incomplete degradation. When plastic is burned, it can produce toxic fumes such as dioxins, which can cause cancer and neurological disorders, as well as high energy consumption during the recycling process [21].

Conventional plastic waste can cause a new problem called microplastics. Microplastics are plastic waste that is larger or smaller than 5 millimeters in size, resulting from ultraviolet degradation. This causes the surface area to become larger and very easy to interact with organic pollutants, and very easy to contaminate zooplankton, making it possible for humans to consume microplastics.

Bioplastics

Bioplastics are currently considered a solution to environmental problems and are expected to replace conventional plastics, at least for certain products [32]. In Table 1 biodegradable plastics are divided into two categories based on their source: biobased and fossil-based. This group of plastics can be classified as environmentally friendly plastics. However, fossil-based plastics result in dependence on fossil fuels [15]. Therefore, plastics made from agro-polymers (agricultural materials) such as starch and cellulose are currently an alternative solution [19].

Bioplastic standards cover various aspects ranging from biodegradability, compostability, renewable content, to food contact safety. Some standards include EN 13432 and ASTM D6400. In Indonesia, bioplastic quality requirements are regulated in the Indonesian National Standard (SNI) 7818:2014 and SNI 7818:2016 [33].

Table 2. Quality requirements for readily degradable plastic bags (SNI : 7818:2014 dan SNI : 7818:2016)

| No | Parameter | Unit | Requirements |
|----|-----------------------|------|----------------------------|
| 1 | Tensile Strength | MPa | Minimum 13.7 (D 882) |
| 2 | Elongation | % | 400-1120 (D 882) |
| 3 | Ease of decomposition | % | >60% for 1 week (ASTM G21) |

Based on Table 2, *tensile* strength is the maximum tensile force that can be obtained before breaking. *Tensile elongation at break* is the maximum increase in film length due to changes in tensile force, measured from the initial length to the point of breakage. The quality of plastic is better if the tensile elongation at break is greater [22]. Biodegradability is the ability of a material, in this case bioplastic, to degrade and decompose naturally in soil, water, or air.

Bioplastic Production

Biodegradable plastic or bioplastic is plastic whose raw materials come from

biomass and can be decomposed with the help of microorganisms [26]. The following are examples of bioplastic applications for food.



Figure 1. Applications of bioplastics for food

The bioplastic in Figure 1 is made from cellulose fermented from butterfly pea flowers. Bioplastics are generally made from natural fibers such as cellulose and plant starch because they are environmentally friendly and biodegradable [20]. The following is the chemical structure of cellulose.

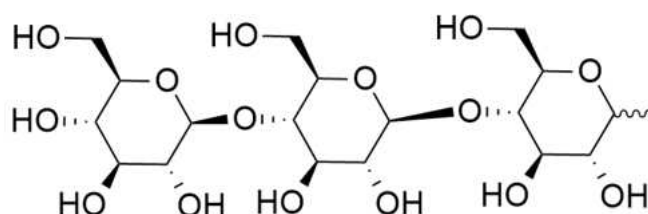


Figure 2. Chemical Structure of Cellulose

Figure 2 shows that cellulose consists of thousands of anhydroglucose groups connected by 1,4- β -glucoside bonds, forming long, linear chain molecules. Thickness, elongation, and *tensile* strength are important physical properties of bioplastics. These physical properties are parameters that greatly influence their intended use for packaging or coating food products [17]. In addition to cellulose and fillers, additional materials are needed in the manufacture of bioplastics to obtain the desired plastic

Plasticizers are added to improve the flexibility and durability of the plastic. The addition of plasticizers can increase the plastic's ability to decompose naturally [18]. Glycerol is a type of plasticizer that is often used in the production of bioplastics. The following is the structure of glycerol.

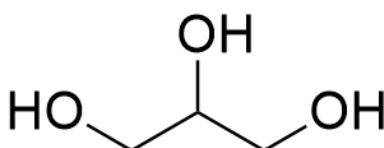


Figure 3. Chemical structure of glycerol

According to Purba et al. [29], glycerol is a *plasticizer* that can increase elongation by reducing the interaction between polymer chain molecules. In addition,

glycerol is easily mixed in film solutions and soluble in water (hydrophilic). Glycerol is also an organic material with a low molecular weight, so it is expected that glycerol can improve the characteristics of the resulting *biodegradable material* [13].

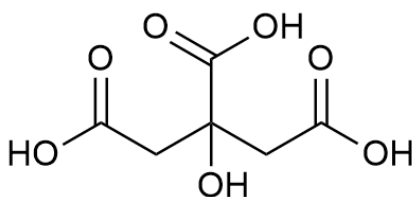


Figure 4. Chemical structure of citric acid

Cross-linking agents are additives that can improve the crystalline structure and optimize the mechanical properties of bioplastics [31]. Citric acid is one of the cross-linking agents that can be used in the bioplastic manufacturing process. Citric acid is more environmentally friendly and more easily degradable [28].

Filler

Fillers are one of the substances added in the bioplastic manufacturing process. Fillers serve to improve the mechanical, physical, and biodegradability properties of bioplastics [23]. Fillers can interact directly with other bioplastic components [30]. Some fillers commonly used in bioplastic manufacturing are chitosan, *carboxymethylcellulose* (CMC), *zinc oxide* (ZnO), and *calcium carbonate* (CaCO₃). These fillers can be adjusted according to the quality of the bioplastic produced. The following is a comparison of several fillers commonly used in the bioplastic manufacturing process.

Table 3 explains the advantages and disadvantages of chitosan, CMC, ZnO, and CaCO₃ as bioplastic fillers. Compared to other fillers, CMC does not have very good thermal stability. However, CMC is soluble in water, which other fillers are not. In addition to improving the mechanical properties of bioplastics, CMC is also safe for use in food. Therefore, CMC is suitable for use as a filler in food packaging bioplastics. The following is the chemical structure of CMC.

The structure of CMC (Carboxy Methyl Cellulose) is a polymer chain consisting of cellulose molecular units and is a linear polymer ether. CMC is an odorless, colorless, non-toxic compound in the form of granules or powder that is soluble in water and has a pH range of 6.5 to 8.0. Each anhydroglucose unit consists of three hydroxyl groups, and carboxymethyl replaces some of the hydrogen atoms from these hydroxyl groups. Carboxymethyl cellulose is added to bioplastics to improve the mechanical properties of the film, provide tensile strength to the polymer film so that the film does not break easily when pulled, and also to smooth the texture of the film. The following is a table of CMC concentration variations for bioplastics.

Table 3. Comparison of Fillers in Bioplastic Production (Source: Rafid et al., 2021 and Ningsih et al., 2019)

| Characteristics | Chitosan | CMC | Zinc Oxide | Calcium Carbonate |
|---------------------|--|---|---|--|
| Chemical formula | $[C_6H_{11}NO_4]_n$ | $C_6H_7O_2(OH)_2OCH_2COO-$ | ZnO | $CaCO_3$ |
| Melting point (°C) | 134,5 | 57 | 1975 | 825 |
| Solubility in water | Insoluble in water | Soluble in water | Not soluble in water | Slightly soluble in water |
| Advantages | Improves the tensile strength and mechanical properties of bioplastics. Has antibacterial properties. | Improves the tensile strength and elongation of bioplastics, Non-toxic and safe for use in food and packaging applications. | Enhances mechanical properties and has antibacterial effects. Improves resistance to UV light. | Improves mechanical properties and can reduce production costs. Provides good thermal stability |
| Disadvantages | Has hydrophobic properties, making it difficult to dissolve in water, which may limit interaction with the matrix. | The extraction and modification process of cellulose into CMC requires energy and chemicals | May cause unwanted chemical reactions with other components in the bioplastic matrix. Potential toxicity if not properly managed in product formulations. | The addition of $CaCO_3$ can cause a decrease in mechanical properties if not distributed properly |

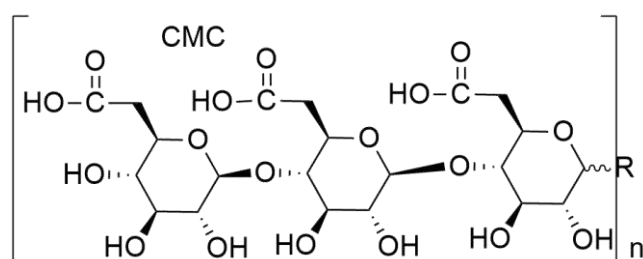
**Figure 5.** Chemical structure of CMC (Carboxy Methyl Cellulose)

Table 4. Effect of CMC Variation on the Thickness and Color of Bioplastics

| No | Main Material | CMC concentration variation | Results |
|----|---|---|---|
| 1. | empty oil palm bunches (Alfiani and Sasria N., 2023) [12] | 1 gram, 2 grams, 3 grams, and 4 grams | The thickness of bioplastic increased with the addition of CMC, with the highest thickness at a CMC concentration of 5 grams with a thickness of 0.125. |
| 2. | Palm oil empty fruit bunches (Zaenab et al., 2023) | 1%, 2%, 3%, and 4% | The higher the concentration of bioplastic color, the more transparent it becomes and the thickness increases. |
| 3. | Cassava starch (Ningsih et al., 2019) [24] | Glycerol 30%, CMC 0%, 3%, 6%, 9%, and 12% | The thickness of the bioplastic increases, with the highest thickness of 0.169 mm at a concentration of 12% |
| 4. | Corn flour (Nurfauzi et al., 2018) [25] | glycerol 2%, CMC 0.5%, 1%, and 1.5% | As CMC concentration increases, thickness increases, with the highest thickness at 1.5% with 0.16 mm |

The table showing variations in CMC concentration indicates that an increase in CMC directly increases the thickness of bioplastics in various base materials. The highest thickness in empty palm oil bunches occurs with the addition of 5 grams of CMC, with a value of 0.125 mm, because hydroxyl groups bind strongly with cellulose when CMC is added, making the bioplastic matrix dense. Another study using similar materials showed that an increase in CMC produces bioplastics with higher thickness and better transparency because the homogeneous distribution of CMC improves the film structure. Cassava starch showed a similar pattern, with thickness continuing to increase when CMC was added until it reached 0.169 mm at a concentration of 12%, caused by amylose starch interacting with CMC through hydrogen bonds to make the film layer denser. The addition of 1.5% CMC to corn flour produced the highest thickness of 0.16 mm because CMC increases the viscosity of the solution and produces a larger solid mass to form a film. These data show that higher CMC concentrations produce bioplastics with greater thickness through increased film formation and molecular bonding in the polymer matrix.

Table 5. Effect of CMC Concentration Variation on Bioplastic Mechanical Strength

| No | Main Ingredient | CMC Concentration Variation | Results |
|----|---|---|--|
| 1. | palm oil empty fruit bunches (Alfiani and Sasria N., 2023) [12] | 1 gram, 2 grams, 3 grams, and 4 grams | The higher the concentration, the higher the tensile strength and elongation values. The highest value was obtained at a CMC concentration of 5 grams at 0.089 MPa |
| 2. | Empty oil palm fruit bunches (Zaenab et al., 2023) | 1%, 2%, 3%, and 4% | As the CMC strength increases, the tensile strength also increases, with the highest tensile strength obtained at a concentration of 4%, namely at 2.103 MPa |
| 3. | Cassava starch (Ningsih et al., 2019) [24] | glycerol 30%, CMC 0%, 3%, 6%, 9%, and 12% | Tensile strength increases with increasing CMC, with the highest tensile strength obtained from a CMC concentration of 9%, which is 0.5281 N/mm ² |
| 4. | Corn flour (Nurfauzi et al., 2018) [25] | Glycerol 2%, CMC 0.5%, 1%, and 1.5% | The highest tensile strength was obtained at a CMC concentration of 1%, which was 8.37 MPa |
| 5. | Jackfruit Seed Starch (Indriani et al., 2023) [16] | 30% starch, 0%, 3%, 6%, 9%, 12%, and 15% of starch mass | Tensile strength did not depend on CMC concentration, but the highest tensile strength was obtained at a concentration of 15% at 2.4549 MPa |
| 6. | nata de pina and rice washing water (Febrianti 2024) [14] | CMC 4, 6, 8, 10.12% glycerol; 4% | The best tensile test was at a CMC concentration of 8% at 0.51 MPa |
| 7. | Green bean starch (Gozali et al., 2020) | 1%, 2%, and 3% | Highest tensile strength at a concentration of 2% was 3.94 MPa |

The tabel of variation in the concentration of CMC shows that the influence of CMC addition to the mechanical properties of bioplastics is very dependent on the type of main material used. In the case of empty fruit bunch, the increase in CMC concentration up to 4% or 4 grams was able to increase the tensile strength due to the hydrogen interaction between cellulose and CMC that strengthened the film structure, although a concentration that is too high may reduce the homogeneity of the matrix. Mountain nagara starch showed an increase in tensile strength along with the addition of CMC with the optimum value at 9% concentration, because the amylose content in starch could bond with CMC through hydrogen bonds so that the structure is denser. For corn starch, the optimum concentration is even low, namely 1% because the content of amylose and amylopectin that is suitable for the number of CMC is limited; the excess addition just disrupts the bonding between starch molecules and lowers the mechanical properties. Meanwhile, in jackfruit seed starch, the variation of CMC did

not significantly affect the tensile strength, although the highest value was recorded at 15% concentration, this is due to the more complex composition of jackfruit seed with protein and crude fiber content. For nata de pina combined with rice washing water, the best result was obtained at CMC concentration of 8%, because the strong structure of bacterial cellulose could be tightened by the addition of CMC at a medium level. Mung bean starch has the optimum value at 2% CMC, where the interaction with amylose is strong enough without making the film too fragile. Generally, it can be concluded that the optimum concentration of CMC varies because of the differences in the chemical composition of the main materials, and broadly speaking low to medium concentrations are effective for starch, while higher concentrations are more suitable for fiber or cellulose-based materials.

Table 6. Effect of CMC Concentration Variation on Bioplastic Degradation

| No | Main Material | CMC Concentration Variation | Results |
|-----|---|---|---|
| 1. | empty oil palm bunches (Alfiani and Sasria N., 2023) [12] | 1 gram, 2 grams, 3 grams, and 4 grams | The higher the concentration, the easier it is to degrade. The highest degradation was at a concentration of 4 grams, namely 22.43% and water absorption of 41.69%. |
| 2. | Empty oil palm bunches (Zaenab et al., 2023) | 1%, 2%, 3%, and 4% | As the CMC concentration increases, the tensile strength also increases, with the highest tensile strength obtained from a concentration of 4%, namely at 2,103 MPa |
| 3. | Cassava starch (Ningsih et al., 2019) [24] | Glycerol 30%, CMC 0%, 3%, 6%, 9%, and 12% | The higher the CMC, the higher the water absorption capacity. Water absorption capacity is directly proportional to degradability. The highest water absorption capacity was obtained at a concentration of 12%, namely 56.604% |
| 4. | Corn flour (Nurfauzi et al., 2018) [25] | Glycerol 2%, CMC 0.5%, 1%, and 1.5% | The higher the CMC concentration, the faster the degradation of mass, which decreases significantly |
| 5. | Jackfruit Seed Starch (Indriani et al., 2023) [16] | 30% starch, 0%, 3%, 6%, 9%, 12%, and 15% of the starch mass | The higher the CMC concentration, the easier it is to degrade and dissolve in water, with the highest degradation at a concentration of 15% at 74.69%. |
| 6.. | nata de pina and rice washing water (Febrianti 2024) [14] | CMC 4, 6, 8, 10, 12% glycerin; 4% | The higher the CMC, the easier it is to degrade. The highest biodegradation is at 35.81% at a concentration of 8%. |

The variation table of CMC concentration shows that the addition of CMC has a different effect on each main bioplastic material due to differences in chemical compositions. In the case of empty fruit bunch, a concentration of 4 grams or 4% not only resulted in the highest degradation (22.43%) but also the strongest tensile (2.103 MPa) as a result of the hydrogen bonding between CMC and cellulose, which both strengthened and accelerated the dissolution.

In the case of ubi nagara starch, an increase in CMC was proportional to water

absorption capacity and degradability, with the highest value at 12% (56.60%) due to the hydrophilic nature of CMC. Corn starch showed optimum results at 1% CMC with the tensile strength of 8.37 MPa, because the excess of CMC just disrupted the bonding of amylose and amylopectin. In jackfruit seed starch, the highest degradation was recorded at 15% (74.69%), indicating that over-addition increased the hydrophilic nature of the film.

Meanwhile, nata de pina with rice washing water reached the best result at 8% with biodegradation of 35.81%, because the bacterial cellulose already had a strong network structure so that the moderate content of CMC was quite effective. In general, low to medium concentrations significantly contributed mechanical strength, while high concentrations were more involved in accelerating bioplastics degradation.

Carboxymethylcellulose (CMC) has a significant impact on the properties of bioplastics, including tensile strength, degradability, and thickness. In general, low to moderate concentrations of CMC can increase tensile strength due to hydrogen bonds that strengthen the matrix structure, while high concentrations accelerate degradation due to the higher hydrophilicity of bioplastics. In addition, the higher the concentration of CMC, the greater the thickness of the bioplastic film, as the amount of film-forming solids increases and the structure becomes denser. However, the optimum concentration differs for each base material, such as 1% for corn flour, 9% for cassava starch, 4% for empty palm fruit bunches, and 8% for nata de pina. Therefore, the selection of CMC concentration must be adjusted to the characteristics of the base material and the application objectives, whether to obtain high mechanical strength, accelerate biodegradation, or produce a certain film thickness. . The interaction between cellulose, CMC, glycerol, and citric acid can occur as shown in the following figure.

Figure 6 above illustrates the reaction between cellulose, CMC, and glycerol. Generally, the reaction between cellulose, CMC, and citric acid is a cross-linking reaction that forms esters. Bacterial cellulose consists of long chains of glucose units linked by β -1,4-glycosidic bonds. Each glucose unit has three hydroxyl groups (-OH) that can interact with *crosslinking* agents and *plasticizers*. One cellulose hydroxyl group will react by binding to one carboxylate group in citric acid or CMC, and the other hydroxyl group will interact with *the plasticizer* through hydrogen bonds [11].

Bioplastics with the addition of CMC are expected to meet the criteria as an environmentally friendly alternative to TNI ration packaging. With the best concentration known, it is hoped that this bioplastic will have optimal mechanical properties for use as food packaging and high biodegradability so that it does not leave traceable residues in the field of operations. This research provides an innovative solution to reduce dependence on plastic while supporting military operations.

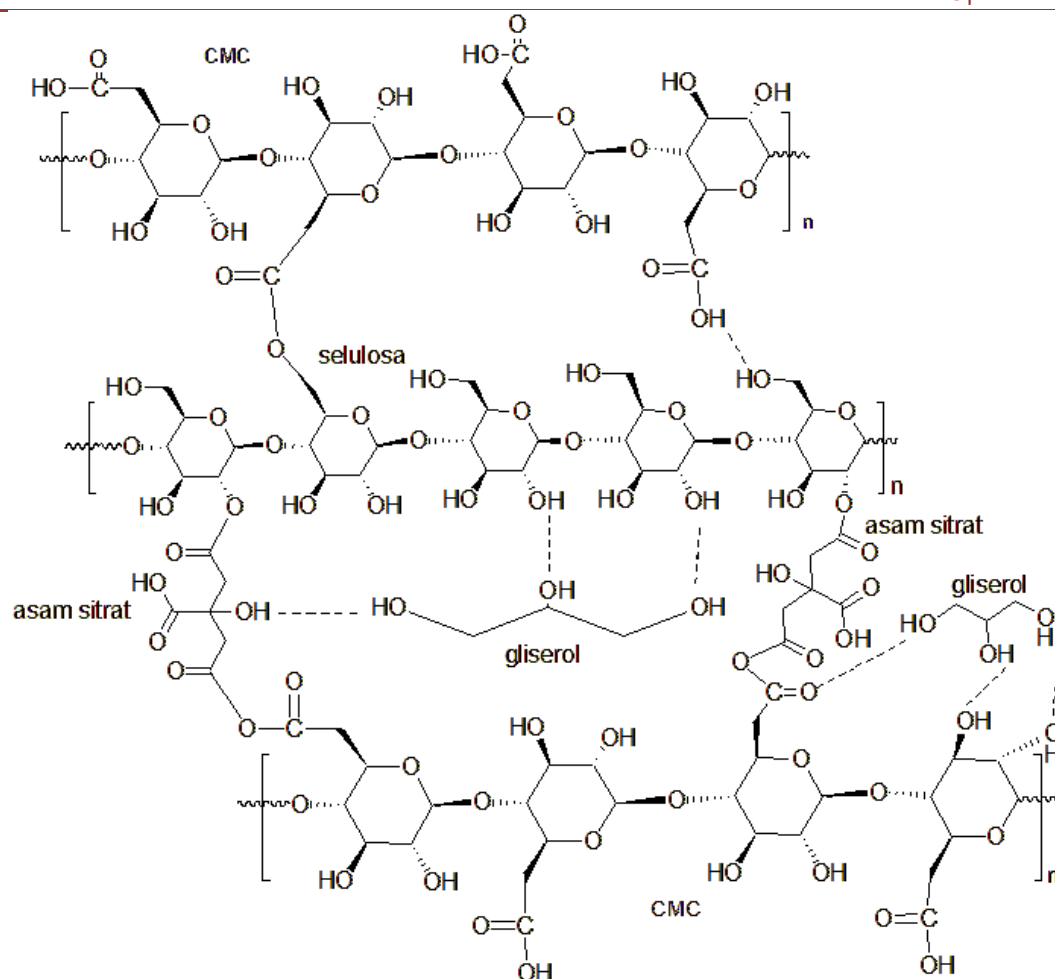


Figure 6. Reaction between cellulose, CMC, citric acid, and glycerol (Aboufotouh et al., 2024).

Conclusion

Literature analysis shows that increasing the CMC concentration generally increases the tensile strength and water absorption of bioplastics, but also accelerates solubility and the rate of biodegradation; as a result, the right formulation is needed to meet the mechanical and durability requirements of the product. Determining the optimal concentration depends on the application objective. Corn starch showed the highest tensile strength at 1% CMC, cassava starch at 9% CMC, and nata de pina at 10–12% CMC. However, if the priority is to accelerate biodegradation, higher concentrations within the tested range are recommended. It is necessary to conduct mechanical testing and further degradation testing under real application conditions before finalizing the formulation.

Acknowledgements

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