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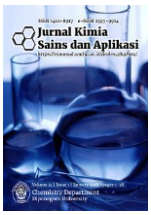

Synthesis and Characterization of Biodegradable Plastics from Areca Nut Shell Cellulose Incorporated Carboxymethyl Cellulose (CMC) and Glycerol

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Abstract

Areca nut shells have a high cellulose content. The potential utilization of areca nut shells as a base material for biodegradable plastics is a key component in the goal of replacing commercial plastics. This study aims to determine the optimal concentration of CMC and glycerol to achieve the best mechanical characteristics of biodegradable plastics. The research method consists of several stages, including the preparation of cellulose from areca nut shells, which involves predelignification and delignification, the synthesis of biodegradable plastic, and the testing of the resulting biodegradable plastic. The mechanical characteristic tests conducted on biodegradable plastics included a tensile strength test (1.27-3.10 MPa), elongation (1.10-1.25%), and Young's modulus (108.54-281.81 MPa) on biodegradable plastics with CMC (4, 5, 6, and 7%) and 4.5% glycerol. In the functional group analysis, biodegradable plastic forms clusters that bond with water, making soil degradation easier. In the thermal analysis, the most significant weight loss occurred between 422.21°C and 492.87°C, which is the stage of cellulose degradation. The swelling value obtained in the areca nut shell cellulose biodegradable plastic is (25.39-11.17%). The use of glycerol affects the value of plastic resistance to water. Estimated degradation times were 45–63 days (3% glycerol), 81–96 days (3.5%), 75–90 days (4%), and 69–84 days (4.5%). Based on ASTM D6400 standards, the material demonstrates biodegradability, with the potential to meet the required degradation thresholds for bioplastics.

1. Introduction

Environmental pollution caused by plastic pollution is a global problem that requires immediate attention and resolution. Two key technologies must be applied from a scientific approach, and global efforts and strategies must be implemented if effective solutions to this problem are to be developed [1]. In 2022, the global trade of plastics reached 436.66 million tons (Mt), of which 111 Mt was attributed to the production of final products [2]. Many items, such as plastic bags and packaging materials, possess a limited functional lifespan but generate a significant volume of environmental waste when discarded. Conventional plastic waste requires a considerable amount of time to decompose, leading to heightened public concern and a growing demand for natural materials. To combat the pollution associated with oil-based plastics, researchers are seeking new materials that offer similar performance at a lower cost.

Films and coatings synthesized from alginate, chitosan, starch, cellulose derivatives, and gelatin have been developed specifically for use in food packaging [3]. Sugar, starch, and cellulose serve as effective substitutes for petroleum-based plastics because they are abundant, eco-friendly, and biodegradable [4]. Biopolymers are increasingly utilized in food packaging due to their unique functional properties. Characteristics such as non-toxicity, accessibility, the ability to interact with biomaterials, and biodegradability are essential factors for industrial consideration. Consequently, biopolymers could replace conventional plastic packaging, which has come under intense environmental scrutiny due to its persistence in the ecosystem [5].

The increasing production of waste and by-products from the agricultural sector has emerged as a significant environmental challenge, contributing substantially to global pollution. A substantial portion of these residues consists of cellulose derived from the complex architecture of plant cell walls [6]. Natural fibers offer numerous advantages in material science, including low density, suitable stiffness, and favorable mechanical properties. Furthermore, plastic materials reinforced with or derived from natural fibers are both recyclable and biodegradable [7]. The integration of cellulose fibers into film production is a common strategy to enhance the structural strength and durability of films [8]. Despite these benefits, cellulose presents challenges due to its highly hydrophilic nature [9]. However, the abundance of hydroxyl groups on the cellulose chain offers a promising platform for chemical modification, allowing researchers to tailor its properties for specific applications [10].

Cellulosic fillers have some drawbacks because of their strong polar character. The most significant issue is their poor compatibility with hydrophobic polymer matrices, resulting in weak interfacial adhesion. Because the final properties of a composite depend on the characteristics of the filler, the matrix, and the interface where they meet, the quality of this bond is critical. A strong interface is necessary for the filler to effectively transfer mechanical loads from the matrix, allowing the resulting material to withstand external forces [11]. The diverse functionalities of pure cellulose and its derivatives enable chemical modifications that introduce stable, permanent functions to the material [12]. One such application involves blending pure cellulose with a Carboxymethyl Cellulose (CMC) matrix to manufacture high-performance biodegradable plastics.

This study explores the fabrication of sustainable bioplastics by incorporating cellulose extracted from areca nut shells into a matrix of CMC and glycerol. As a derivative of natural cellulose, CMC is highly valued for its tailored hydrophilicity, viscous stability, and robust mechanical performance [13]. The areca shells utilized in this research are sourced from North Aceh, Indonesia, where agricultural processing generates massive quantities of underutilized lignocellulosic biomass annually. Within this composite system, glycerol serves as a critical plasticizing agent, disrupting internal hydrogen bonding to enhance the flexibility of the resulting films [12].

The efficacy of CMC as a base for environmental materials is supported by several specialized studies. Cheng *et al.* [12] demonstrated that while blending CMC with washed cottonseed meal (CSM) produced effective biodegradable films, there was a clear trade-off: increasing the CSM ratio reduced water permeability but compromised the overall tensile strength. This suggests that the CMC concentration remains a primary determinant of structural integrity.

Innovation in bioactive packaging was further advanced by Łopusiewicz *et al.* [14], who modified CMC matrices with fungal melanin and carvacrol. Their results showed that although carvacrol increased film opacity, the combination provided superior antioxidant protection and mechanical durability, positioning these composites as viable alternatives to synthetic food wraps.

Song and Othman [15] observed that adding lignin to CMC-palm fiber films improved thermal stability and moisture resistance, yet SEM analysis indicated a lack of uniform distribution within the matrix. Conversely, Bátori *et al.* [16] addressed similar homogeneity issues in citrus-based films by introducing maleic anhydride (MA). Rather than acting strictly as a cross-linker, the MA functioned as a compatibilizer, eliminating structural voids (holes) and significantly improving the adhesion between the cellulosic fibers and the pectin-based matrix. These findings underscore the importance of chemical additives in improving the performance of plastics derived from agricultural waste.

The areca nut shell serves as a significant source of wood fiber due to its high cellulose and lignin content. As a lignocellulosic resource, these shells are rich in cellulose, hemicellulose, and lignin, offering mechanical and thermal properties that make them ideal for biocomposites and sustainable packaging [17]. Chemically, the areca nut consists of polyphenols (10%-30%), polysaccharides (18%-25%), fibers (10%-15%), fatty acids (10%-15%), and alkaloids (0.3%-0.7%) [18] [19]. Detailed fiber analysis reveals a composition of 53.20% alpha-cellulose, 32.98% hemicellulose, 7.20% lignin, 0.64% fats and waxes, and 1.05% ash [19].

These fibers are characterized by high inherent strength and thermal resistance, positioning them as an effective alternative to synthetic reinforcements in polymer matrices. Utilizing areca nut shell fibers enables the repurposing of agricultural waste into eco-friendly materials, thereby reducing reliance on hazardous synthetic fibers [20]. The current research investigates a novel formulation combining areca shell cellulose with CMC and glycerol. To date, there is no documented literature exploring this specific ternary mixture as a soluble, biodegradable plastic.

CMC, a versatile cellulose derivative, acts as a functional filler that promotes chemical crosslinking and enhances the structural properties of thin films. It is widely recognized for its biocompatibility, non-toxicity, and water solubility, which have led to its extensive application in the fields of pharmacology and biology [21]. In this study, CMC concentrations were varied at 4%, 5%, 6%, and 7%, while glycerol (as plasticizer) was tested at

levels of 3%, 3.5%, 4%, and 4.5%. These specific ranges were selected based on preliminary trials to optimize the physical performance of the material. The experimental design aims to determine how varying these components influences the mechanical strength, thermal stability, functional group characteristics, water resistance, and biodegradation rates of the resulting areca-based bioplastics.

2. Experimental

2.1. Materials and Research Methodology

Areca nut shells were obtained from plantations in the Sawang District, North Aceh, and processed to produce cellulose fibers. Distilled water was supplied by PT Bratachem (Surabaya, Indonesia). Sodium hydroxide (NaOH, pro analysis, Merck, 1.06498.1000), glycerol (85%, pro analysis, Merck, 1.04094.0500), and carboxymethyl cellulose (CMC, food grade, Wealthy–Merck) were used in this study. Polypropylene (PP, CAS No. 9003-07-0), maleic anhydride (MA, 99%, Sigma-Aldrich, 8.00408.1000), and xylene (Merck Supelco, 108297) were also employed.

This study utilized cellulose derived from areca nut shells and CMC–glycerol as the main components. The research methodology consisted of several stages, including cellulose preparation from areca nut shell fibers, extraction through pre-delignification and delignification processes, synthesis of biodegradable plastic, and characterization of the resulting plastic materials.

2.2. Preparation of Cellulose from Areca Nut Shell

A total of 100 g of areca nut shells were thoroughly washed and air-dried for 24 hours. The dried shells were then ground into a fine powder using a blender and sieved through a 120-mesh screen to obtain a uniform particle size.

2.3. Areca Nut Shell Extraction

The extraction process consisted of two stages: predelignification and delignification. In the predelignification stage, areca nut shell powder (in varying weights) was mixed with distilled water at a ratio of 1:10 (w/v). The mixture was stirred thoroughly and refluxed at 100°C for 3 hours. The resulting suspension was filtered, and the solid residue was oven-dried at 100°C for 120 minutes. In the delignification stage, the predelignified powder was soaked in a 10% NaOH solution until it was fully immersed, then autoclaved at 121°C for 60 minutes [22]. The mixture was filtered to obtain solid residues, which were subsequently treated with 3.5% sodium hypochlorite (NaOCl) solution (w/v) mixed with distilled water at a 1:1 ratio. The suspension was boiled for 10 minutes, filtered, and washed thoroughly with distilled water. Finally, the solids were oven-dried at 100°C for 60 minutes, resulting in pure cellulose powder (alpha-cellulose) with a purity of 90%.

2.4. Synthesis of Biodegradable Plastics

Biodegradable plastic specimens were prepared by mixing cellulose, glycerol, CMC, and PP through a grafting process at 125°C. The formulations consisted of varying CMC concentrations (4%, 5%, 6%, and 7%) and glycerol concentrations (3%, 3.5%, 4%, and 4.5%). In the grafting process, 25 g of polypropylene was dissolved in 100 mL of xylene, followed by the addition of maleic anhydride (1% of the xylene volume) and benzoyl peroxide (0.1% of the maleic anhydride weight). The mixture was stirred at 125°C until homogeneous, producing PP-g-MA. Separately, 50 g of cellulose was combined with CMC and glycerol (12.5 g each), then mixed with PP-g-MA. The blend was heated and stirred until it was uniform, followed by solvent evaporation until a white, dough-like product was obtained. The resulting material was molded using a hot-press machine at 165°C under a pressure of 1 psi for 10 minutes, with a holding time of 8 minutes. The molded specimens were cooled to room temperature before further testing.

2.5. Characterization and Testing

The mechanical properties of biodegradable plastics derived from areca nut shell cellulose, blended with glycerol and CMC, were evaluated in terms of tensile strength, elongation at break, and elastic modulus. The tests were conducted in accordance with ASTM D638-14 (Type II, suitable for materials with thicknesses between 7 and 14 mm) using a Universal Testing Machine [23]. The tensile strength (σ) was calculated using Equation (1), where σ is the tensile strength (MPa), F_{max} is the maximum force applied (N), and A is the cross-sectional area of the specimen under tensile stress (mm^2).

$$\sigma = \frac{F_{max}}{A} \quad (1)$$

Elongation at break (ε) was determined using Equation (2), where ε represents the strain, l is the final gauge length after elongation (mm), and l_0 is the initial gauge length (mm).

$$\varepsilon = \frac{l - l_0}{l_0} \quad (2)$$

Fourier Transform Infrared Spectroscopy (FTIR) was employed to identify functional groups in the biodegradable plastic samples. The analysis was performed using a Shimadzu 8400S spectrometer (Japan), with spectra collected in the range of 550–4000 cm^{-1}. FTIR spectroscopy is a method that uses infrared light to identify molecular bonds and groups of atoms in different substances. The results from the spectrum are the result of the absorption and transmission of molecules. The molecules of a sample form the spectrum results. This technique provides information about the molecular structure of the material and the presence of characteristic chemical bonds.

Thermal stability was examined using Thermogravimetric Analysis (TGA) with a TGA50 model instrument (Heating Rate: 10°C/min; Gas Flow: 20–50 mL/min; and range (25–600°C). TGA determines the resistance of biodegradable plastics to thermal degradation by measuring changes in mass as a function of temperature.

The swelling behavior of the plastics was assessed according to ASTM D2765-16 [24]. The specimens were

initially weighed and then immersed in the solvent for 24 hours at 25°C. After immersion, the swollen samples were removed, the surface solvent was gently wiped off, and the samples were reweighed. The specimens were subsequently dried to a constant weight and weighed again to determine the final mass. The degree of swelling was calculated using Equation (3), where the swelling degree is expressed as the percentage increase in mass relative to the initial weight.

$$Swelling\ degree = \frac{W_s - W_0}{W_0} \times 100\% \quad (3)$$

Where, W_s is the weight of the swollen sample, and W_0 is the initial dry weight.

Soil burial degradation was tested following ASTM D5988 [25]. Plastic specimens (5 × 2 cm) were weighed (M_0), buried 30 cm deep in soil, and retrieved after four days, with a period of 16 days. The temperature and pH levels employed in soil burial degradation analyses (32°C and 5.5–7) are widely regarded as conducive to facilitating biodegradation. Notably, the specific parameters for soil moisture during the burial process for biodegradable plastics depend on the prevailing environmental conditions. The samples were cleaned, dried, and reweighed (M_1). The biodegradation percentage was calculated using Equation (4), where M_0 is the initial mass (g), and M_1 is the final mass (g).

$$\text{Biodegradabillity (\%)} = \frac{M_0 - M_1}{M_0} \times 100\% \quad (4)$$

3. Results and Discussion

3.1. Mechanical Properties: Tensile Strength, Elongation, and Young's Modulus

Texture analysis was used to assess the material's mechanical properties. Tensile strength has been demonstrated to exhibit an inverse correlation with elongation rate, with Young's modulus demonstrating a comparable response. The mechanical test results of areca nut shell cellulose–based biodegradable plastics plasticized with glycerol and modified with CMC are summarized in Table 1. The tensile strength values of areca nut shell cellulose-based biodegradable plastics with glycerol and CMC ranged from 1.27 to 3.10 MPa. In this formulation, glycerol functioned as a plasticizer, while CMC acted as the matrix component. The highest tensile strength was achieved at a 7% CMC concentration with 4.5% glycerol. These results indicate that increasing the CMC content enhances the tensile strength of biodegradable plastics.

Glycerol, a molecule of modest size and flexibility, has been shown to disrupt the hydrogen bonds that facilitate the interconnection of the CMC polymer chains. The occurrence of this rupture has been demonstrated to result in a decrease in the stiffness of the polymer matrix. This decrease, in turn, has been shown to allow the chains to move more freely past each other under stress. Glycerol is a versatile chemical that can exist in various forms. Glycerol has been shown to disrupt the hydrogen bonds that bind CMC polymer chains [26]. The introduction of glycerol to CMC results in an interaction with the polymer chains, leading to the disruption of the hydrogen bonds that bind neighboring CMC polymer molecules. However, the tensile strength values obtained in this study cannot be directly compared with those of conventional plastics such as HDPE (11–25 MPa) and PP (15–45 MPa), which are reported in the MatWeb Material Property dataset under the classification of Polypropylene, Extrusion Grade.

Comparable findings were reported by Tavares *et al.* [27], who observed improved moisture barrier properties and higher tensile strength in corn- and cassava-based films containing 50% w/w CMC. The superior performance of CMC-based films relative to carboxymethyl starch (CMS) films was attributed to the higher molecular weight of cellulose compared to starch derivatives [28]. Based on these results, further optimization may require a higher CMC content to achieve stronger biodegradable plastics.

Elongation, defined as the extent to which a material stretches under tensile load, reflects its elasticity. Plastics with high elongation values typically exhibit greater creep. In this study, the values of elongation for areca nut shell biodegradable plastics with 4.5% glycerol and 7% CMC ranged from 1.10% to 1.25%. The enhancement in flexibility was attributable to the presence of a plasticizer with a smaller molecular size. This plasticizer enabled the molecules to traverse the interstices between polymer chains, thereby diminishing the strength of hydrogen bonds between molecules. Consequently, this reduction in bond strength facilitated greater molecular mobility [29]. Although glycerol as a plasticizer is known to enhance elongation and water vapor permeability in wheat films [30], the low elongation values obtained here suggest limited flexibility. These values are considerably lower than those of PET, which range between 15% and 165% [31], and comparison with CMC is Young's modulus (E) = 0.053 ± 0.017 MPa; stress (σ) = 0.136 ± 0.016 MPa; and strain (ε) at break = 621.38 ± 58.28%.

Table 1. Mechanical properties of areca nut shell cellulose–based biodegradable plastics containing CMC and glycerol

Glycerol (%)	CMC (%)	Tensile strength (MPa)	Elongation (%)	Young's Modulus (MPa)
4.5	4	1.27	1.17	108.54
	5	2.46	1.25	196.80
	6	2.76	1.19	231.93
	7	3.10	1.10	281.81
Pure PP [32]	0	32.63±0.17	88.59±60.14	464.43±12.39
Areca nut shell [19]	0	147-322	10.23-13.15	1124±3155

The reduction in tensile strength may result from weak interfacial bonding between fibers in the composite, which tends to decrease as fiber volume fraction increases [33]. A similar trend was observed in CMC films prepared by *Mimosa pigra* peel, where elongation increased 1.25-fold as the glycerol content rose from 10% to 30%, while the tensile strength declined from 10 MPa to 5 MPa [34]. This phenomenon is linked to the disruption of hydrogen bonding by glycerol, which increases chain mobility and intermolecular spacing [35]. The elastic modulus of a material reflects its stiffness and is inversely related to its elongation. It provides a quantitative measure of a material's resistance to deformation under stress and is determined by the ratio of stress to strain.

In this study, the elastic modulus values of areca nut shell cellulose-based biodegradable plastics ranged from 108.54 to 281.81 MPa, with the highest value observed at a 7% CMC concentration. For comparison, the MatWeb Material Property dataset reports modulus values for polypropylene (extrusion grade) in the range of 0.680–3.60 GPa, with elongation values between 8% and 750% [36]. These values are significantly higher than those obtained for the areca nut shell-based plastics, indicating that the experimental films are far less stiff and flexible compared to conventional commercial plastics.

The stiffness and flexibility of CMC-based films are strongly influenced by glycerol concentration. Low glycerol content results in stiffer films with adequate flexibility, suitable for applications requiring structural integrity, whereas higher glycerol levels increase flexibility but reduce tensile strength, which is desirable for pliable applications such as food packaging [37]. Film thickness and environmental conditions also play a significant role; thicker films generally require more glycerol to maintain flexibility, while thinner films require less to prevent breakage. Under high humidity, increasing glycerol concentration can prevent brittleness by counteracting moisture-induced stiffness. Thus, tailoring glycerol concentration enables optimization of CMC films for different application requirements. Glycerol can migrate to the surface of the polysaccharide or the material it is in contact with. Glycerol forms a secondary bond with the polymer, and its small size allows it to diffuse to the polymer's surface or leach into the water. As a result of this plasticizer loss, the polymer becomes brittle with poor mechanical properties [38].

3.2. Chemical Properties with Fourier Transform Infrared (FTIR) Analysis

The objective of this analysis was to identify the functional groups present in the biodegradable plastic samples using FTIR. Each sample was placed in a designated holder, and its infrared spectrum was recorded to determine the characteristic absorption bands. The resulting spectra display the relationship between wave number and absorbance intensity, which reflects the molecular vibrations of the functional groups present. Figure 1 presents the FTIR spectrum of the biodegradable plastic sample prepared with 7% CMC and 4.5% glycerol.

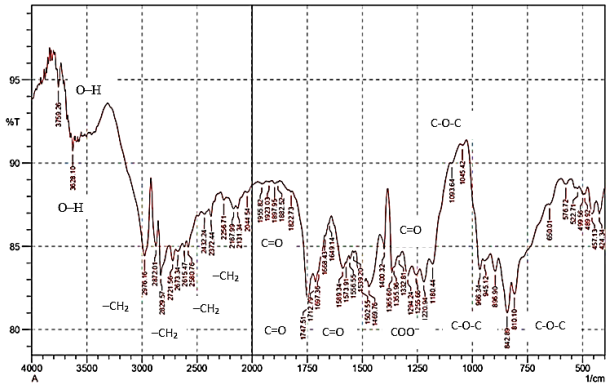


Figure 1. FTIR spectra of areca nut shell cellulose–based biodegradable plastics

The FTIR spectra of areca nut shell cellulose–based biodegradable plastic are presented in Figure 1. The spectra were normalized at 872 cm^{-1}, which corresponds to the β-D-glycosidic linkage, a distinctive feature of cellulose materials [39]. In the sample, C–O–C stretching vibrations of areca nut shell cellulose incorporated in the biodegradable plastic were observed at 1045.42 cm^{-1} and 1093.64 cm^{-1}, while additional C–O–C vibrations in β-D-glycosidic linkages appeared at 810.10 cm^{-1}, 842.89 cm^{-1}, and 896.90 cm^{-1}. These findings are consistent with a previous study by Zhao *et al.* [40], which reported characteristic vibrations at 1161 cm^{-1} for C–O–C stretching, 1022 cm^{-1} and 1030 cm^{-1} for C–O stretching, and 896 cm^{-1} for the C–O–C β-D-glycosidic linkage.

Figure 1 shows the FTIR spectra of the areca nut shell–based biodegradable plastic blended with CMC and glycerol. The presence of $-CH_2$ and C=O stretching vibrations, corresponding to methylene groups and carbonyl esters, was observed at 2976.16, 2872.01, 2829.57, and 1712.79 cm^{-1}, which characterize organic compounds of maleic anhydride and PP, indicating the role of PP–g–MA as a compatibilizer. For comparison, pure LDPE films typically exhibit symmetric and asymmetric $-CH_2$ peaks at 2847 and 2925 cm^{-1} [41], while PE–g–MA shows characteristic peaks at 2914 and 2847 cm^{-1} for polyethylene and 1705 cm^{-1} and 718 cm^{-1} for the C=O group of maleic anhydride [42].

Additional features included sharper bands between 800 and 1150 cm^{-1} with increasing glycerol concentration, while an absorption at 925 cm^{-1} was detected only in glycerol-plasticized films, suggesting the presence of enhanced hydroxyl groups in the polymer matrix. Spectra of CMC revealed bands at 1589.34–1400.32 cm^{-1}, corresponding to the antisymmetric and symmetric stretching of carboxylate (COO^-) groups, indicating substitution of carboxymethyl groups on the cellulose backbone [43].

Characteristic O–H likely residual moisture or free –OH bands stretching was observed at 3350.3 cm^{-1} (unbleached cellulose from palm bunches), and at 3628.10 and 3759.26 cm^{-1} in the biodegradable plastic. Peaks at 1641.4 cm^{-1} and 1649.14 cm^{-1} were assigned to the O–H bending vibrations of water molecules and cellulose from biodegradable plastic derived from areca nut shells, consistent with earlier studies [43, 44]. Overall, the FTIR

results demonstrate the presence of organic and hydrophilic clusters, which enhance the film's interaction with water and facilitate biodegradation in soil [45].

3.3. Thermal Properties using Thermogravimetric Analysis (TGA)

The thermal stability of the biodegradable plastic containing 7% CMC and 4.5% glycerol was analyzed by TGA, as shown in Figure 2, which illustrates mass changes as a function of temperature. An initial, gradual weight loss, starting at around 30°C, is attributed to residual impurities and additives. A major weight loss occurred between 422.21 and 492.87 °C, corresponding to cellulose thermal degradation and subsequent crystallization. Under these conditions, the bulk of the material underwent decomposition, with complete mass depletion occurring at approximately 600°C (initial sample weight: 20 mg, residual weight: 98.954%). These results align with the reported degradation profile of lignocellulosic fibers, which typically occurs between 400°C and 500°C [46].

Cellulose bioplastics exhibit relatively high thermal resistance. Significant weight loss occurs between 230°C and 370°C, as the amorphous regions decompose at lower temperatures than crystalline cellulose. Further degradation between 370°C and 600°C results in the formation of polycyclic aromatic ring compounds, observed as char, the solid byproduct of thermal decomposition [47, 48]. However, previous studies have reported that the incorporation of glycerol reduces the thermal stability of cellulose films, as glycerol promotes water absorption within cellulose fibers. The absorbed water disrupts fiber interactions, thereby diminishing film stability at elevated temperatures [49].

3.4. Water Absorption using Swelling Degree Analysis

The water resistance of the biodegradable plastics was evaluated through swelling tests, where the samples were immersed in water. The degree of swelling represents the amount of water absorbed by the material and provides an indication of its hydrophilic or hydrophobic character. The swelling test results for areca nut shell cellulose–based biodegradable plastics containing 4, 5, 6, and 7% CMC with glycerol contents of 3, 3.5, 4, and 4.5% are presented in Figure 3.

Figure 3 presents the swelling values of areca nut shell cellulose-based biodegradable plastics containing CMC and glycerol, ranging from 11.17% to 25.39%. The results indicate that glycerol concentration has a significant influence on the water resistance of the plastics. A higher glycerol content increased the swelling value, indicating lower water resistance, whereas a lower glycerol content produced a reduced swelling value. The maximum swelling value of 25.39% was achieved with a 4.5% glycerol and 4% CMC solution. Glycerol contains multiple –OH groups, which promote hydrogen bonding with water molecules, thereby enhancing the water absorption capacity of the bioplastics [50].

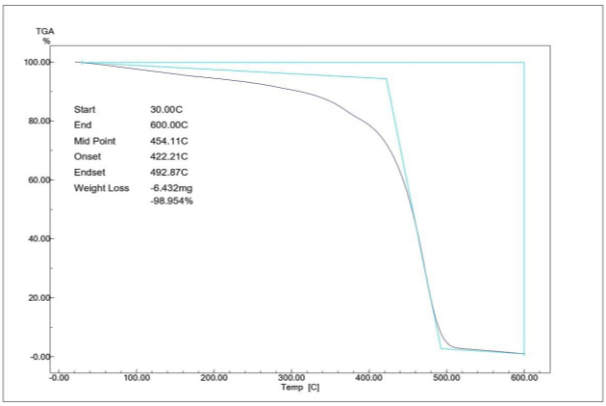


Figure 2. TGA thermogram of areca nut shell cellulose-based biodegradable plastics

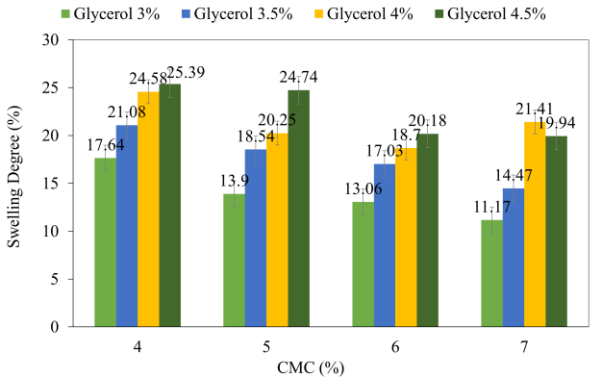


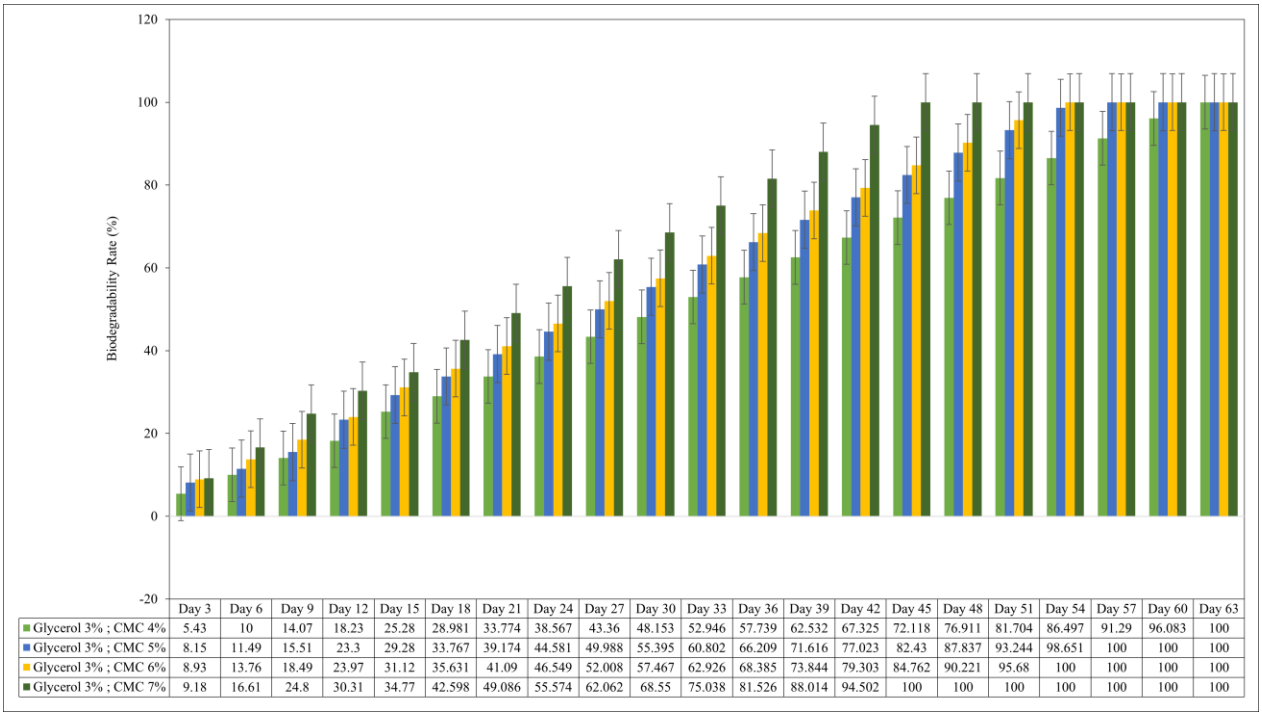
Figure 3. Swelling degree of areca nut shell cellulose-based biodegradable plastics with CMC and glycerol

Obasi and Igwe [51] reported similar findings in yam starch-based polymers. Their study compared polymers blended with polypropylene compatibilizer grafted with maleic anhydride (PP-g-MA) against blends without a compatibilizer. The results demonstrated that the incorporation of PP-g-MA improved water resistance, as blends lacking PP-g-MA exhibited ~28% higher water absorption. Likewise, Shrestha *et al.* [33] developed starch-based bioplastics with glycerol and polyvinyl alcohol (PVA) plasticizers, showing that increased starch content led to greater water uptake in the starch/PVA blends. The higher starch-based bioplastics degraded more rapidly but exhibited stronger resistance to acid and base for up to 45 and 42 hours, respectively.

According to the Food and Agriculture Organization (FAO), the safe moisture content limit for biodegradable products is 12%. According to the Food and Agriculture Organization (FAO), the safe water content limit for biodegradable products is 12%. In this study, only samples of CMC variations processed without 7% CMC and 3% glycerol approached this limit [52]. On the other hand, hydrophilic plasticizers such as glycerol can also diminish the barrier properties of the films, as they contain hydroxyl groups capable of forming hydrogen bonds with water, thereby increasing the hydrophilicity of the plasticized film. This increased hydrophilicity is responsible for the increased water vapor permeability of the films, which is undesirable for packaging applications [38].

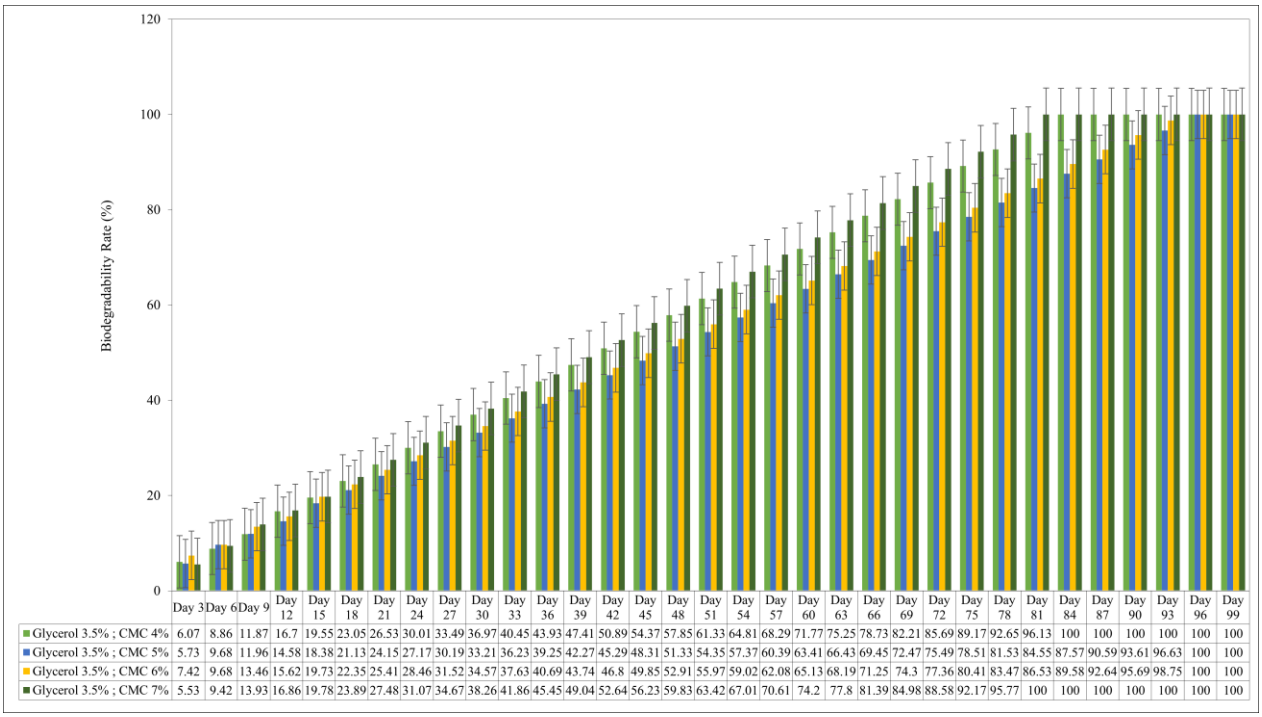
Table 2. Biodegradability rate of areca nut shell cellulose-based biodegradable plastics with CMC and glycerol under soil burial conditions

Glycerol (%)	CMC (%)	n-Days Mass Loss				
		3	6	9	12	16
3	4	5.43	10.00	14.07	18.23	25.28
	5	8.15	11.49	15.51	23.21	29.82
	6	8.93	13.76	18.49	23.97	31.12
	7	9.18	13.61	24.80	30.12	34.77
3.5	4	6.07	8.86	11.87	16.70	19.55
	5	5.73	9.68	11.96	14.58	18.38
	6	7.42	9.68	13.46	15.62	19.73
	7	5.53	9.42	13.93	16.86	19.78
4	4	6.73	9.38	13.21	15.70	19.84
	5	6.67	10.57	13.79	17.58	19.48
	6	4.59	8.15	12.20	17.71	20.28
	7	6.09	8.88	12.75	14.35	20.05
4.5	4	5.51	8.63	12.39	5.83	19.48
	5	4.17	8.06	11.86	16.20	19.39
	6	6.26	9.39	13.46	18.10	21.02
	7	5.12	8.43	13.11	17.15	22.60



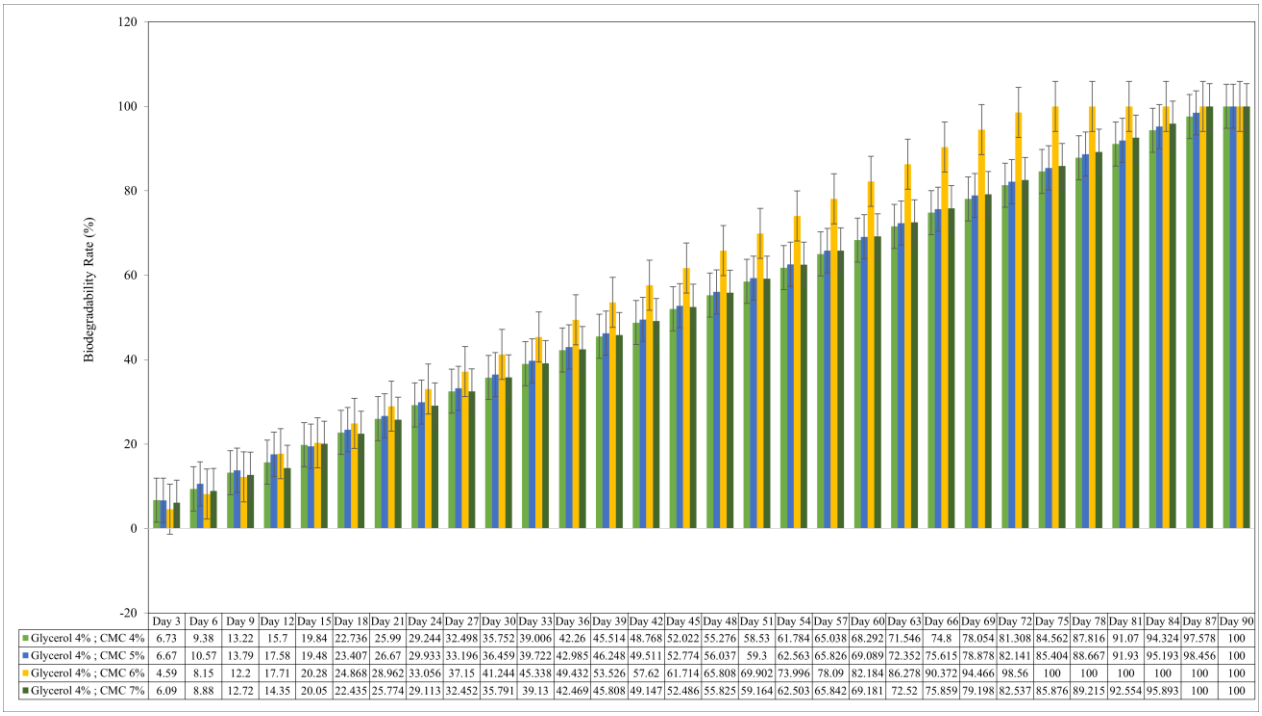
	Day 3	Day 6	Day 9	Day 12	Day 15	Day 18	Day 21	Day 24	Day 27	Day 30	Day 33	Day 36	Day 39	Day 42	Day 45	Day 48	Day 51	Day 54	Day 57	Day 60	Day 63
Glycerol 3% ; CMC 4%	5.43	10	14.07	18.23	25.28	28.981	33.774	38.567	43.36	48.153	52.946	57.739	62.532	67.325	72.118	76.911	81.704	86.497	91.29	96.083	100
Glycerol 3% ; CMC 5%	8.15	11.49	15.51	23.3	29.28	33.767	39.174	44.581	49.988	55.395	60.802	66.209	71.616	77.023	82.43	87.837	93.244	98.651	100	100	100
Glycerol 3% ; CMC 6%	8.93	13.76	18.49	23.97	31.12	35.631	41.09	46.549	52.008	57.467	62.926	68.385	73.844	79.303	84.762	90.221	95.68	100	100	100	100
Glycerol 3% ; CMC 7%	9.18	16.61	24.8	30.31	34.77	42.598	49.086	55.574	62.062	68.55	75.038	81.526	88.014	94.502	100	100	100	100	100	100	100

Figure 4. Extrapolation rate on biodegradable plastics for the glycerol 3% estimation of total plastic decomposition



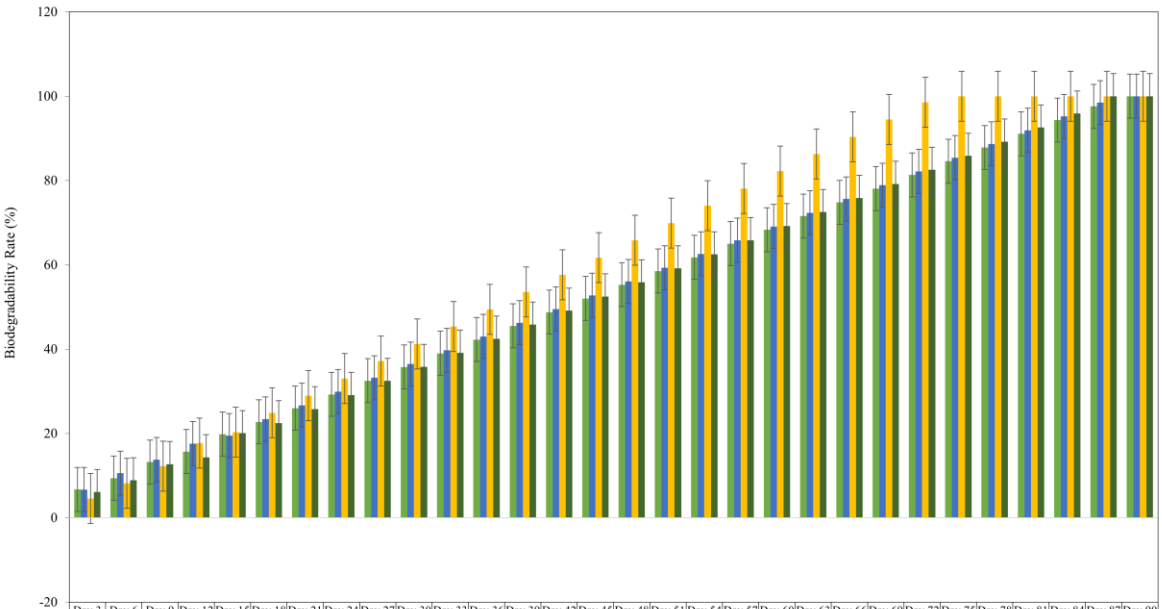
	Day 3	Day 6	Day 9	Day 12	Day 15	Day 18	Day 21	Day 24	Day 27	Day 30	Day 33	Day 36	Day 39	Day 42	Day 45	Day 48	Day 51	Day 54	Day 57	Day 60	Day 63	Day 66	Day 69	Day 72	Day 75	Day 78	Day 81	Day 84	Day 87	Day 90	Day 93	Day 96	Day 99
Glycerol 3.5% ; CMC 4%	6.07	8.86	11.87	16.7	19.55	23.05	26.53	30.01	33.49	36.97	40.45	43.93	47.41	50.89	54.37	57.85	61.33	64.81	68.29	71.77	75.25	78.73	82.21	85.69	89.17	92.65	96.13	100	100	100	100	100	100
Glycerol 3.5% ; CMC 5%	5.73	9.68	11.96	14.58	18.38	21.13	24.15	27.17	30.19	33.21	36.23	39.25	42.27	45.29	48.31	51.33	54.35	57.37	60.39	63.41	66.43	69.45	72.47	75.49	78.51	81.53	84.55	87.57	90.59	93.61	96.63	100	100
Glycerol 3.5% ; CMC 6%	7.42	9.68	13.46	15.62	19.73	22.35	25.41	28.46	31.52	34.57	37.63	40.69	43.74	46.8	49.85	52.91	55.97	59.02	62.08	65.13	68.19	71.25	74.3	77.36	80.41	83.47	86.53	89.58	92.64	95.69	98.75	100	100
Glycerol 3.5% ; CMC 7%	5.53	9.42	13.93	16.86	19.78	23.89	27.48	31.07	34.67	38.26	41.86	45.45	49.04	52.64	56.23	59.83	63.42	67.01	70.61	74.2	77.8	81.39	84.98	88.58	92.17	95.77	100	100	100	100	100	100	100

Figure 5. Extrapolation rate on biodegradable plastics for glycerol, 3.5% estimation of total plastic decomposition



	Day 3	Day 6	Day 9	Day 12	Day 15	Day 18	Day 21	Day 24	Day 27	Day 30	Day 33	Day 36	Day 39	Day 42	Day 45	Day 48	Day 51	Day 54	Day 57	Day 60	Day 63	Day 66	Day 69	Day 72	Day 75	Day 78	Day 81	Day 84	Day 87	Day 90
Glycerol 4% ; CMC 4%	6.73	9.38	13.22	15.7	19.84	22.736	25.99	29.244	32.498	35.752	39.006	42.26	45.514	48.768	52.022	55.276	58.53	61.784	65.038	68.292	71.546	74.8	78.054	81.308	84.562	87.816	91.07	94.324	97.578	100
Glycerol 4% ; CMC 5%	6.67	10.57	13.79	17.58	19.48	23.407	26.67	29.933	33.196	36.459	39.722	42.985	46.248	49.511	52.774	56.037	59.3	62.563	65.826	69.089	72.352	75.615	78.878	82.141	85.404	88.667	91.93	95.193	98.456	100
Glycerol 4% ; CMC 6%	4.59	8.15	12.2	17.71	20.28	24.868	28.962	33.056	37.15	41.244	45.338	49.432	53.526	57.62	61.714	65.808	69.902	73.996	78.09	82.184	86.278	90.372	94.466	98.56	100	100	100	100	100	100
Glycerol 4% ; CMC 7%	6.09	8.88	12.72	14.35	20.05	22.435	25.774	29.113	32.452	35.791	39.13	42.469	45.808	49.147	52.486	55.825	59.164	62.503	65.842	69.181	72.52	75.859	79.198	82.537	85.876	89.215	92.554	95.893	100	100

Figure 6. Extrapolation rate on biodegradable plastics for the glycerol 4% estimation of total plastic decomposition



	Day 3	Day 6	Day 9	Day 12	Day 15	Day 18	Day 21	Day 24	Day 27	Day 30	Day 33	Day 36	Day 39	Day 42	Day 45	Day 48	Day 51	Day 54	Day 57	Day 60	Day 63	Day 66	Day 69	Day 72	Day 75	Day 78	Day 81	Day 84	Day 87	Day 90
Glycerol 4% ; CMC 4%	6.73	9.38	13.22	15.7	19.84	22.736	25.99	29.244	32.498	35.752	39.006	42.26	45.514	48.768	52.022	55.276	58.53	61.784	65.038	68.292	71.546	74.8	78.054	81.308	84.562	87.816	91.07	94.324	97.578	100
Glycerol 4% ; CMC 5%	6.67	10.57	13.79	17.58	19.48	23.407	26.67	29.933	33.196	36.459	39.722	42.985	46.248	49.511	52.774	56.037	59.3	62.563	65.826	69.089	72.352	75.615	78.878	82.141	85.404	88.667	91.93	95.193	98.456	100
Glycerol 4% ; CMC 6%	4.59	8.15	12.2	17.71	20.28	24.868	28.962	33.056	37.15	41.244	45.338	49.432	53.526	57.62	61.714	65.808	69.902	73.996	78.09	82.184	86.278	90.372	94.466	98.56	100	100	100	100	100	100
Glycerol 4% ; CMC 7%	6.09	8.88	12.72	14.35	20.05	22.435	25.774	29.113	32.452	35.791	39.13	42.469	45.808	49.147	52.486	55.825	59.164	62.503	65.842	69.181	72.52	75.859	79.198	82.537	85.876	89.215	92.554	95.893	100	100

Figure 7. Extrapolation rate on biodegradable plastics for glycerol, 4.5% estimation of total plastic decomposition

3.5. Biodegradability Rate with Soil Burial Analysis

The biodegradability of areca nut shell cellulose–based biodegradable plastics was evaluated through a soil burial test to assess their decomposition rate under microbial activity. The degradation behavior of samples with varying CMC and glycerol concentrations, along with extrapolated degradation rates used to estimate total decomposition time, is presented in Table 2. The degradation time ranged from 19.39 to 34.77 days, depending on the plastic composition. Based on extrapolation of the degradation curves, complete degradation (100%) occurred most rapidly at approximately 45 days for samples containing 3% glycerol and 7% CMC.

Further extrapolation results (Figure 4) indicate that samples with 3% glycerol and CMC contents of 4–7% reached full degradation within 45–63 days. For samples containing 3.5% glycerol (Figure 5), the estimated degradation time increased to 81–96 days, while samples with 4% glycerol (Figure 6) and 4.5% glycerol (Figure 7) showed estimated degradation times of 75–90 days and 69–84 days, respectively. These results demonstrate that glycerol concentration significantly influences the biodegradation rate, with higher glycerol contents generally promoting faster degradation due to increased polymer chain mobility and water uptake.

In addition to plasticizer content, biodegradation is significantly influenced by external factors, including microbial activity, temperature, soil moisture, and environmental conditions. CMC, a cellulose derivative, enhances interfacial interactions within the polymer matrix, improving film-forming properties while maintaining biodegradability.

Comparable findings have been reported in the literature. Zahri *et al.* [53] demonstrated that a consortium of native Antarctic bacteria was able to degrade 94.42% of waste canola oil (WCO) and 86.83% of virgin canola oil (PCO) within seven days, with temperature and substrate concentration identified as significant factors influencing the biodegradation process. In comparison, the areca nut shell cellulose–based biodegradable plastics incorporating glycerol and CMC exhibited biodegradability rates that comply with the ASTM D5988 standard, which requires a minimum of 60% biodegradation within six months for homopolymers or mixed copolymers, and 90% for copolymers and polymer blends [54]. Similarly, Steven *et al.* [39] reported that cellulose-based bioplastics exhibited degradation exceeding 40% within five days, indicating the high biodegradability potential of cellulose-derived materials.

Overall, the biodegradation behavior of areca nut shell cellulose–based biodegradable plastics incorporating glycerol and CMC satisfies the ASTM D5988 standard, which requires at least 60% biodegradation within six months for homopolymers or mixed copolymers and 90% for polymer blends [55]. These results confirm the suitability of areca nut shell cellulose as a promising raw material for biodegradable plastic applications.

4. Conclusion

In this research, glycerol functioned as a plasticizer and CMC as the matrix. The highest tensile strength was achieved with 7% CMC and 4.5% glycerol, indicating that increasing CMC concentration enhances the tensile strength of biodegradable plastics. FTIR analysis of the areca nut shell-based plastics with glycerol and CMC confirmed the presence of organic and hydrophilic clusters, which facilitate water binding and, consequently, soil degradation. Thermogravimetric analysis demonstrated that an increase in mass loss

following decomposition corresponded to higher thermal resistance. The addition of glycerol was also found to influence water resistance: higher glycerol concentrations improved plastic resistance to water, while lower concentrations reduced it. The material becomes more hydrophilic, or attracted to water, when it absorbs more water. This reduces its ability to resist penetration or damage from water. The biodegradation time of areca nut shell cellulose-based plastics with glycerol and CMC variations ranged from 19.39 to 34.77 days. In addition to the environmental benefits of this approach, the utilization of areca nut shell waste has the potential to provide socio-economic benefits for communities involved in areca nut cultivation and trade. However, the utilization of this waste is currently limited, and further research is needed to explore the potential economic benefits for these communities in the future.

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