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# Composite cassava starch/chitosan/Pineapple Leaf Fiber (PALF)/Zinc Oxide (ZnO): Bioplastics with high mechanical properties and faster degradation in soil and seawater

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ABSTRACT

Biopolymers based on starch, cellulose and proteins extracted directly from biomass with or without modification have been widely used as the natural resources to produce biodegradable plastic. Starch has several disadvantages: strong hydrophilic behavior and inferior mechanical properties when compared with synthetic polymers. Starch is also mostly soluble in water and will be decomposed before undergoing the gelatinization process. To provide resistance and mechanical strength of starch, several fillers (reinforcement) in the form of metal and natural materials are usually added to the polymer matrix. Zinc oxide (ZnO) nanoparticle and natural fiber as a lightweight material that is biocompatible, nontoxic, cost-effective and exhibit strong antibacterial activity can be considered as a reinforcement of starch-based bioplastic. The present study, the reinforcing effect of ZnO on the mechanical, antibacterial, and physical properties of bioplastic films in the form of cassava starch/chitosan/pineapple leaf fiber (PALF)/ZnO. The highest value of elongation at break is for 16 % ZnO-bioplastics which could be completely decomposed only 21 days in ordinary soil and only 18 days in seawater. The packaging tests using slice bread showed antimicrobial properties with no fungal growth for 30 days of bioplastic coatings with 10, 13, and 16 % ZnO NPs. The results in this study indicated that, the ZnO and PALF plays an important role in reinforcing the physical, mechanical, and antibacterial properties of starch/chitosan/PALF-based bioplastic.

## 1. Introduction

Bioplastic is an innovation that has proven to be an environmental-friendly alternative with potential to replace conventional plastics because it's biodegradable, sustainable, and renewable. Bioplastics can be synthesized from natural organic materials such as polysaccharides, proteins, and lipids, but starch-based bioplastics are the most promising because of their abundance in nature [1]. However, starch-based materials showed low water barrier characteristics and poor mechanical performance compared to the non-natural polymers due to high hydrophilicity and affinity to water, such disadvantages severely restrict their widespread application. Some ways to overcome this deficiency are mixing with other biopolymers, modifying starch, biowaste [2], nanoparticles [3–7] and additional natural fiber [8–12].

Natural fibers have recently attracted the attention because of the advantages that these fibers provide over conventional reinforcement materials, and the development of natural fiber composites has been a subject of interest for the past few years. These natural fibers are low-cost fibers with low density and high specific properties. These are biodegradable and non-abrasive, unlike other reinforcing fibers. Also, the natural fibers are renewable resources, thus providing a better solution of sustainable supply and has low cost, low density, least processing expenditure, no health hazards and good mechanical and physical properties [13].

Pineapple leaf fiber (PALF) is a plant fiber that is very hydrophilic and has a relatively good potential as a filler due to its high cellulose content but lower hemicellulose and lignin content [14]. The fiber component consists of 8[illegible] % cellulose, 12 % lignin, and several properties such as a density of 1.44 g/cm$^3$, Young's modulus of 82 GPa, tensile strength of 180 MPa, and 3.2 % strain which has the potential as a composite reinforcement [15]. Therefore, pineapple leaf fiber has higher mechanical properties compared to other natural fibers [14]. Thus, the addition of PALF in the starch matrix has great potential as a renewable and biodegradable polymer.

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In recent years, the incorporation of nanoparticles (NPs) into composite materials has attracted a lot of attention. Composite materials containing nanoparticles can produce high-performance and innovative materials. Inorganic nanoparticles maintain interfacial interactions in bioplastics and significantly improve the mechanical properties of the matrix. According to the literature, all mechanical and thermal properties of bioplastic composites were improved by the addition of inorganic nanoparticles [4]. Zinc oxide (ZnO) nanoparticles have been attracting great attention as a nanofiller since it shows good biocompatibility, it is harmless and environmentally friendly. Its crystalline structure, very large surface area and excellent mechanical properties also become advantages of ZnO to reinforce polymeric nanocomposites. Since it has high mechanical properties, the good interaction between ZnO and polymers would generate better transfer of ZnO mechanical properties to polymer matrices [15]. Furthermore, ZnO is also reported as multifunctional nanofiller due to its antimicrobial activity. The antibacterial activity of ZnO is due to the production of reactive oxygen species that generates the oxidation of bacterial cells cytoplasm, resulting in cell death [16,17]. The ZnO also shows barrier activity against enzymatic degradation, so it is very suitable for food packaging applications. In our recently publication for composite cassava starch/chitosan/ZnO successfully shows improve shelf life as a food packaging [18], we continue in this study by additional PALF in the form of composite cassava starch/chitosan/PALF/ZnO for environmentally friendly concept. The degradation performance of composite cassava starch/chitosan/ZnO reported in Ref [18] showed 21 days buried in soil and need 21 days in sea water due to low elongation at break. By additional PALF for environmentally concept, the composite in this study should be high elongation at break and faster degradation in any media.

Therefore, in the present study, PALF and ZnO was used to enhance the performance of starch/chitosan-based bioplastics. PALF is used for increasing degradation performance (faster in any media) and ZnO for increasing shelf life of food. The reinforcing effect of various concentration of ZnO on the physical properties, structure, and biodegradation properties of starch/chitosan/PALF-based bioplastic was investigated. This is expected the composite in this study could be high potentials to be applied for mass production as food packaging in future with faster degradation and high shelf life of food.

## 2. Materials and methods

### 2.1. Materials

Cassava starch, chitosan, pineapple leaf fiber (PALF) (see preparation in the next section), powder chitosan pharmaceutical grade 98 % extracted from shrimp shell with particle size: mesh 100–300, moisture 8 %, and residue of ignition 0.75 %, bulk density 0.15–0.3 g/cm$^3$, low molecular weight, viscosity 20–300 cP (1 wt% in 1 % acetic acid at 25 °C,) and soluble in dilute aqueous acid produced by CV. Multiguna Indonesia. Glycerin ($C_3H_8O_3$) (molecular weight 92.09, boiling point 290 °C) (Merck), acetic acid ($CH_3COOH$) with concentration of 5 % (Merck) (purity 99.8 %, density 1.05 g/cm$^3$, Boiling Point 118 °C, Melting Point 16. 6 °C), sulfuric acid ($H_2SO_4$) solution p.a. ≥25 % with concentration 1 Molar (Merck), ZnO powder with maximum particle size 45-micron, purity 99.999 % (Merck), and aquades.

### 2.2. Pineapple Leaf Fiber preparation

Bioplastic film preparation begins with the manufacture of pineapple leaf fiber (PALF). First, the pineapple leaves are cleaned and cut to a size of 20 × 20 cm$^2$, then heated for 2 h at 100 °C. After that, the pineapple leaves are wrapped in aluminum foil for 24 h. In the next stage, the pineapple leaves are cut, and the fiber is taken using a cutter. Furthermore, the fibers obtained were soaked in NaOH solution (1 Molar) for 60 min, then washed using water. After cleaning, the fibers were heated for 15 min at 70 °C in the microwave [9].

### 2.3. Bio-composite sample preparations

Bioplastic film preparation in this study using the casting method. First, 20 ml distilled water was mixture in acetic acid/glycerin/starch with weight ratio of 2/4/6 into a beaker and add 3 % of ZnO, then homogenized using a magnetic stirrer to form a gel. Furthermore, 8 % chitosan (of the total weight of starch) was dissolved in sulfuric acid and mixed into the starch-bioplastic gel, then homogenized again. Pineapple leaf fiber was put into the middle of the bioplastic gel, after that put into a mold, and heated using a microwave for 14 h covered with aluminum foil. The bioplastic preparation was repeated for (6,10,13,16) % of ZnO. The namely samples for Chitosan/ZnO/PALF for various amount of ZnO: 3 %, 6 %, 10 %, 13 %, and 16 % are respectively: PN1, PN2, PN3, PN4, and PN5. The cassava starch/chitosan/PALF/ZnO bioplastic film preparation method can be seen in Fig. 1.

### 2.4. Bioplastic film sample testing preparation

The X-Ray diffraction (XRD) was used for analyzing the structural properties, (Shimadzu 7000 X-ray diffraction) with CuKα radiation (λ = 1.5405 Å) for 2θ from 5° to 60°, operates at 40 kV and 30 mA. All samples were cut in circles with a diameter of 1.5 cm. The absorption capacity of bioplastic samples was obtained by immersing the sample in water for 1 h, then weighed and soaked again for 1 h. This cycle lasts until it reaches saturation. The absorption capacity of bioplastics is formulated by Eq. (1) [19]. The samples were planted in 2 types: ordinary soil and seawater for duration of 7 days, 14 days, and 21 days. After the stockpiling period the initial weight and weight after stockpiling were measured and the percentage of biodegradation calculated by the Eq. (2) [19]. Packaging Test was carried out to see the feasibility of the bioplastic film for food packaging. The bioplastic film is made with a size of 10 × 10 cm$^2$, then 6 slices of white bread with a size of 2 × 2 cm$^2$ are wrapped using bioplastic films. This test was carried out for 30 days [3] with some modification from reference to see the resistance of the bread.

$$\text{Water Absorption } (\%) = \frac{m_1 - m_0}{m_0} \times 100\% \tag{1}$$

$$\text{Biodegradation } (\%) = \frac{m_0 - m_1}{m_0} \times 100\% \tag{2}$$

where $m_0$ and $m_1$ are the initial and final masses, respectively.

## 3. Result and discussion

### 3.1. Water absorption test

Fig. 2 (a) shows the results of the absorption test for 7 h which every hour was measured for all PN samples with the addition of chitosan, PALF, and various ZnO concentration. The higher concentration of ZnO (PN 1-PN 5), the lower the percentage of bioplastic absorption to water [7]. Research of [20] stated that ZnO acts as a nanofiller that functions to cover the pores of bioplastics so that bioplastics with the addition of ZnO are very difficult to absorb water.

The bioplastic samples were saturated when entering the 3rd hour immersion and after the 7th hour would experience a saturation phase. Thus, for the variation of PN samples, the resistance of bioplastics to water absorption only lasted for 7 h. The addition of PALF into bioplastics causes an increase in the percentage of water absorption even though there are hydrophobic chitosan and ZnO [21]. The maximum value for the PN 1 sample is 66.94 % and continues to increase in absorption as the concentration of ZnO increases. This is because PALF is a highly hydrophilic plant fiber and has a relatively good potential as a filler/reinforcement material due to its high cellulose content [14]. Research of [8] stated that starch and cellulose have similar

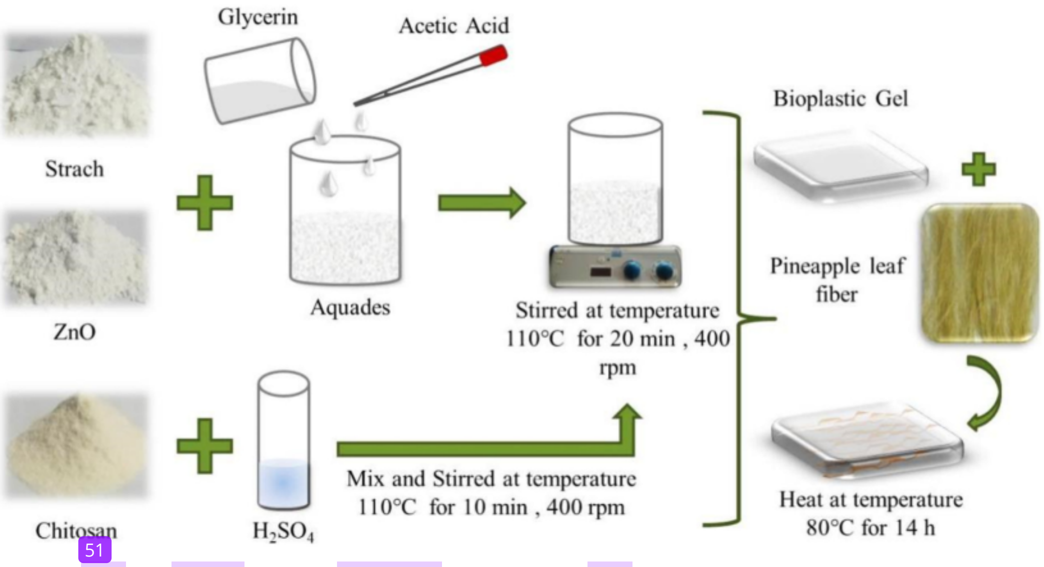


**Fig. 1.** The schematic illustration of preparation composite cassava starch/chitosan/pineapple leaf fiber (PALF)/ZnO.

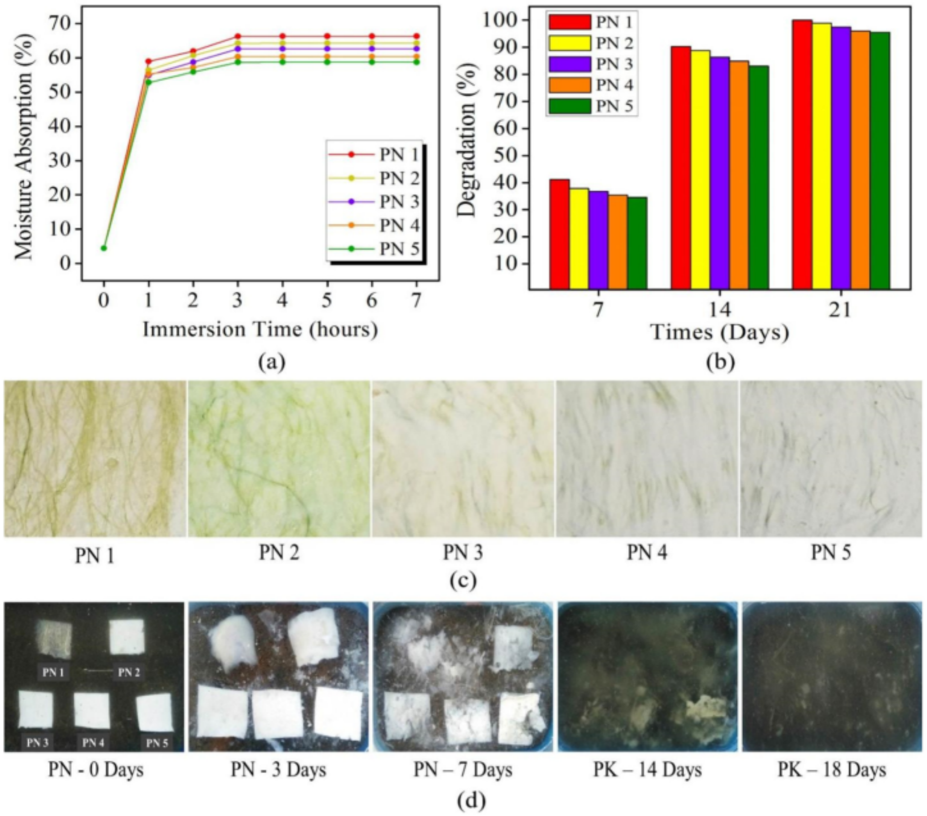


**Fig. 2.** (a) The water absorption percentage of PN Samples in this study. (b) Percentage degradation of PN samples in ordinary soil, (c) optical photograph view of bioplastics PN samples for various amount of ZnO. The vertical line indicated the PALF in composite cassava starch/chitosan/pineapple leaf fiber (PALF)/ZnO, and (d) The photograph visual seawater degradation test of PN samples for 0 days, 3 days, 7 days, 14 days, and 18 days.

polysaccharide structures, fillers in the form of reinforcing pineapple leaves, are fibers with a high level of cellulose and will occupy the starch matrix, thereby increasing water absorption. Similarly, research by [22]. The higher cellulose content of natural fibers provides a good mechanical advantage but increases hydrophilicity. This happens because hydrogen bonds in cellulose molecules tend to form intra-molecular hydrogen bonds including with water molecules. Thus, it can be concluded that the addition of natural fibers into the starch/chitosan/ZnO matrix, can increase the hydrophobicity and mechanical properties of bioplastics.

Bioplastics are degraded in the environment in a very short time probably due to the presence of C═C and C—H bonds as the effect of PALF which also contributes to the mechanical properties [9]. On the 14th days, the percentage of PN 1-PN 5 degradation was 90.23 %, 88.78 %, 86.35 %, 84.94 %, and 83.00 %, and for the 21st days, the percentage of PN 1-PN 5 degradation was 100 %, 98.94 %, 96.06 %, and 95.51 %, respectively. These results prove that the presence of PALF can accelerate the degradation process because the constituent materials are natural materials that can be decomposed by soil microbes or in other words, are hydroscopic [22]. This is consistently based on the water absorption test in Fig. 3(a), that the maximum value obtained by PN 1 is 66.94 % and continues to increase in absorption with increasing ZnO concentration. This is because PALF is a highly hydrophilic plant fiber and has a relatively good potential as a filler/reinforcement material due to its high cellulose content [14]. Table 1 shows the tensile strength, biodegradation performance, and application of chitosan/ZnO and cellulose/ZnO with and without additional materials. The composite chitosan/cellulose/ZnO with additional acetate phthalate shows degraded 30–50 % in 28 days although tensile strength of 10.53–16.9 MPa [23]. For chitosan/ZnO with additional polypropylene, the tensile strength of 22.59–31.35 MPa with weight loss 17.82 % after 3 months in soil [24] and by additional neem oil shows more high tensile strength 30–60 MPa but less weight loss after 5 days.

### 3.2. Soil degradation test

Fig. 2(b) shows the percentage of bioplastic degradation of PN samples for 7, 14, and 21 days. The bioplastic sample was added with pineapple leaf fiber which serves to improve the mechanical properties of the bioplastic and is expected to accelerate the degradation process. Based on the results obtained, the percentage value of PN 1-PN 5 degradation on the 7th day was 41.19 %, 37.86 %, 36.79 %, 35.40 %, and 34.49 %. The results show that there is a decrease in the value of degradation along with an increase in the concentration of ZnO. This is because ZnO has hydrophobic properties so that the absorption of water from the soil into the bioplastic matrix will be reduced.

Fig. 2 (c) shows the bioplastic produced for the PN 1 in the form of a thick, brownish-colored sheet that was transparent but not see-through, elastic, and had small pores. As the concentration of ZnO increases, the resulting bioplastics ranging from PN 2 – PN 5 are stiffer, the color of the sample becomes whiter, and there are almost no pores in the bioplastic. ZnO acts as a nanofiller that functions to cover the pores of the starch-bioplastics matrix [18,20,43] so that bioplastics containing ZnO tend to be stiffer. The green color in all bioplastics in this study come from the PALF used. (Jamiluddin et al., 2018) revealed that the addition of PALF in the starch-bioplastic matrix can improve the mechanical properties of bioplastics [44].

According to research by (Mittal et al., 2019) that natural fiber-reinforced composites showed the least biodegradability in soil. In addition, the total weight loss (%) in the Coir-Epoxy composite was 51.1 % lower than that of the pure PALF-Epoxy composite [45]. This result is attributed to the presence of high cellulose content in PALF which results in easy and fast absorption of water molecules. The mechanism of biodegradation involves penetration of water molecules, breaking of strong covalent bonds, and breakdown of hemicellulose & cellulose by the action of microorganisms. And according to research (Mutmainna et al., 2019) using corn starch/chitosan with various in the weight of PALF (3 %, 6 %, and 9 %) as reinforcement showed that bioplastic decomposed about 45 % on day 3 for PALF 9 %, and 14th about 80 % [9]. Compared with 6 % PALF, the percentage of bioplastic degradation on the 3rd day was only around 35 %, and on the 14th day was only 70 %. Bioplastics are degraded in the environment in a very short time probably due to the presence of C═C and C—H bonds as an effect of PALF [9]. Thus, it was concluded that the addition of PALF could increase the percentage of degradation in the soil even though there was a concentration of ZnO in the bioplastic samples. The processes degradation of bioplastic in soil starts with incoming energy from the sun. The energy from the sun will increase temperature of the soil and at night the temperature of the soil decrease (increase humidity) which can make bioplastic stress. In a day more oxygen produced and at night more $H_2O$ which absorbed in the bioplastics. These $H_2O$ and $O_2$ will break the bond when they are getting energy from the sun and produce $OH^-$ and $O^-$ ion [24]. The ZnO in the bioplastics will produce electron ($e^-$)–hole ($h^+$) pairs when the energy from the sun incoming to the samples. The ($h^+$) will react with $H_2O$ molecules produce $OH^-$ and $H^+$, and ($e^-$) will react with $O_2$ produce $O^-$ and $O_2^-$. These ions more reactive and attach in hydrogen bond of the starch and chitosan and continue breaking another atomic bonding that makes decreasing molecular weight, decreasing strength, and decreasing weight, and finally fragmentation to form small part of bioplastics. The simpler mechanism of biodegradation is involving penetration of water molecules, breaking of strong covalent bonds, and breakdown of hemicellulose and cellulose [45]. These processes will be changing chemical, mechanical, and physical properties of the bioplastics until the last products are $CH_2 + CO_2 + H_2O$ + biomass

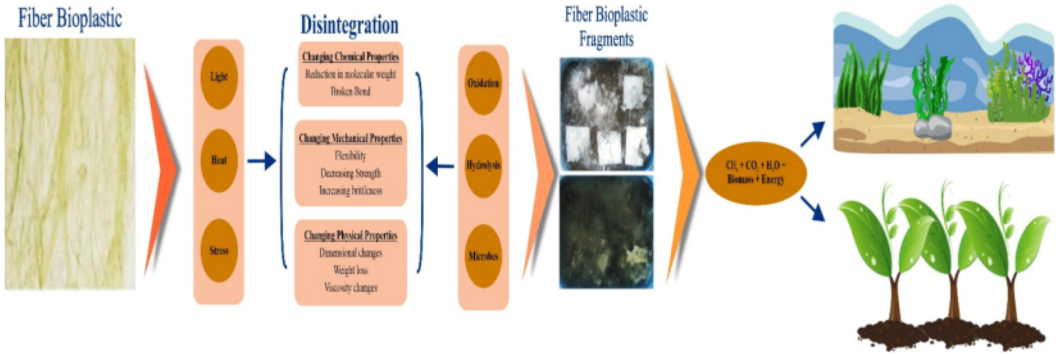


**Fig. 3.** Schematic illustration how the process of disintegration and then fragmentation bioplastics caused by the light, heat, stress, oxidation, hydrolysis, and microbes with the final product is $CH_2 + CO_2 + H_2O$ + biomass and energy as a nutrient to grow the plant.

**Table 1**
Tensile strength, biodegradation performance, and application of biopolymer chitosan/ZnO and cellulose/ZnO with and without additional material from various references [a-t].

| Materials | Additional materials | Tensile strength (Mpa) | Biodegradation | Applications | Refs. |
|---|---|---|---|---|---|
| Chitosan and ZnO | | 5.56–31.9 | | Packaging | [25] |
| Chitosan/Cellulose and ZnO | Acetate phthalate | 10.53–16.9 | Biodegraded about 30–50 % in 28 days. | Vegetables and fruits packaging film | [23] |
| Chitosan and ZnO | Neem oil | 30–60 | Less weight loss after 5 days | Food packaging | [26] |
| | Polypropylene | 22.59–31.35 | The weight loss 17.82 % after 3 months in soil | Bioplastic | [24] |
| | | 43.9–80.2 | | Medical materials | [27] |
| | | 12.84–41.73 | | Biopolymer | [28] |
| | PVA | 19.76–24.23 | | Hydrophilic wound and burn dressings | [29] |
| | Poly Lactid Acid (PLA), $TiO_2$ | 1.716–5.039 | | Bioplastic | [30] |
| | Banana Starch, glycerol | 2.5–36 | Completely biodegraded 120 min | Bioplastic | [31] |
| | | 4.11–12.79 | | Antimicrobial | [32] |
| Cellulose and ZnO | | 59.2–202.0 | | Antibacterial | [33] |
| | Poly (lactic acid) | 32.22–45.28 | | Food packaging | [34] |
| | Ag | 36.6–46.4 | | Catalyst and antibacterial | [35] |
| | grapefruit seed extract | 7.69–21.01 | | Wound healing and antibacterial | [36] |
| | | 55 | | Flexible electronics | [37] |
| | | 39.0–55.0 | 36 % (4 weeks buried in soil) | Food packaging | [38] |
| | | 44.3–92.4 | | Nano paper for food packaging | [39] |
| | Natural rubber | 12. 5–16.34 | | Multifunctional polymer | [40] |
| | Oregano essential oil | 40.27–68.11 | | Green packaging | [41] |
| | *Dioscorea opposita* mucilage and glycerol | 4.79–21.60 | | Food packaging | [42] |

(residue) and energy.

### 3.3. Seawater degradation test

Degradation testing is carried out to determine the level of bioplastic degradation in seawater. The degradation test is parameter to determine the environmentally friendly nature of the bioplastic [46]. The test was carried out for 1–18 days with visual observations in bioplastics every day.

Based on the observations, Fig. 2(d) shows that, on the 3rd day, bioplastics underwent a wider change in shape, and some bioplastics decomposed in each sample. This is because the resistance of the sample to absorb water has reached the saturation phase. Based on Fig. 3(a) it shows that, the bioplastic reaches saturation when it enters a 3-h immersion and when the immersion is >7 h, the bioplastic will reach the saturation phase, which causes a change in the shape of the sample. Based on the previous explanation, PALF is a highly hydrophilic plant fiber and has a relatively good potential as a filler/reinforcement material due to its high cellulose content [14]. The high cellulose content in pineapple leaf fiber results in easy and fast absorption of water molecules.

On the third day, the PN 1–PN 5 samples were degraded (about 30 %), which was indicated by the presence of small parts of the bioplastic samples that were released from the matrix. On the 7th day (degraded about 70 %), PN 1 samples degraded faster which was indicated by the release of fibers from the starch matrix and the decomposition of all parts of the sample compared to PN 2-PN 5 this was due to the small concentration of hydrophobic ZnO [7]. On the 14th day (degraded about 100 %), PN 1 and PN 2 samples completely decomposed. While the PN 3 sample still contained some parts of the bioplastic that had not been decomposed but with a very small size (about 1 %). Likewise, for samples PN 4 and PN 5 which only lost half of the bioplastic but have been degraded, which is characterized by the thinning of the bioplastic sample layer and yellowish color, this is also influenced by the concentration of ZnO in each sample. Then on the 18th day, all samples were completely degraded but there were still small fragments of the bioplastic matrix and some PALF. Based on the explanation above, it can be concluded that PALF could accelerate the degradation process even though there are chitosan and ZnO in the matrix-bioplastics. The seawater is containing NaCl and $H_2O$ molecule. The energy from the sun will increase the temperature of seawater and then breaking the bond of NaCl and $H_2O$ molecule which produce ion $OH^-$ and $Cl^-$. From the inside of the bioplastics, there is ZnO will produce electron ($e^-$)–hole ($h^+$) pairs when the electromagnetic (UV and visible light) incoming to the samples. The holes will react with $H_2O$ molecules from the seawater and split into $OH^-$ and $H^+$ ions [18]. These ions more reactive which attach in hydrogen atom of the starch and chitosan and continue breaking another atomic bonding that makes the bioplastics stress. These processes will changing chemical properties consequently reduce molecular weight, changing mechanical properties by decreasing strength and increasing brittleness, and changing physical properties by dimension changes and weight loss.

Fig. 3 shows how these phenomena degradation of bioplastics to produce $CH_2 + CO_2 + H_2O$ + biomass and energy as a nutrient to grow well the plant. The biggest contribution in bio-deterioration by disintegration process of the bioplastics come from the sun, the light of the sun brings the energy, these energies will produce heat, continue with oxidation, and hydrolysis that make the bioplastics stress, but the time of these process strongly depend on the humidity, temperature, and oxygen content [43]. All these processes will be changing chemical properties by breaking the bond consequently reduce molecular weight, changing mechanical properties by decreasing strength and increasing brittleness, and changing physical properties by dimension changes and weight loss [18,43]. From the soil, microorganisms (bacteria, archaea, fungi, and protozoa) were contributed to disintegration by bio-fragmentation, this process occurs by breaking down polymers into oligomer and monomer [43]. The disintegration by bio-deterioration and bio-fragmentation continue during the process of degradation with the last product is $CH_2 + CO_2 + H_2O$ + biomass (residue) and energy, where the residue of bioplastics generally nontoxic, so can be consumed by living organism in soil or in ocean.

Fig. 4 (a) shows Fourier transform infra-red (FTIR) spectra of starch/chitosan/PALF indicated by 1 from our previous published data [9] and we have added composite chitosan/ZnO taken from Ref. [25] and chitosan/cellulose/ZnO (CCZ) from Ref. [23]. The OH vibration bond clearly can be seen in wavenumber 3200–3500 $cm^{-1}$ [47]. Fig. 4 (b)

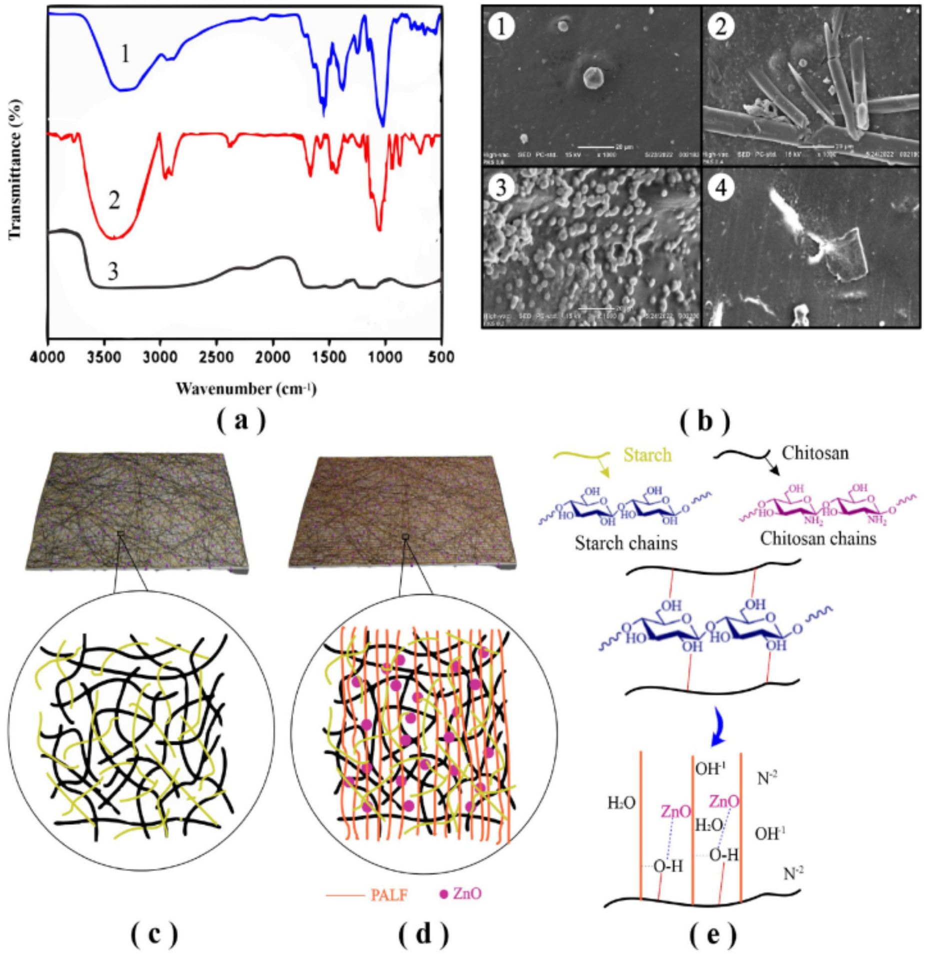


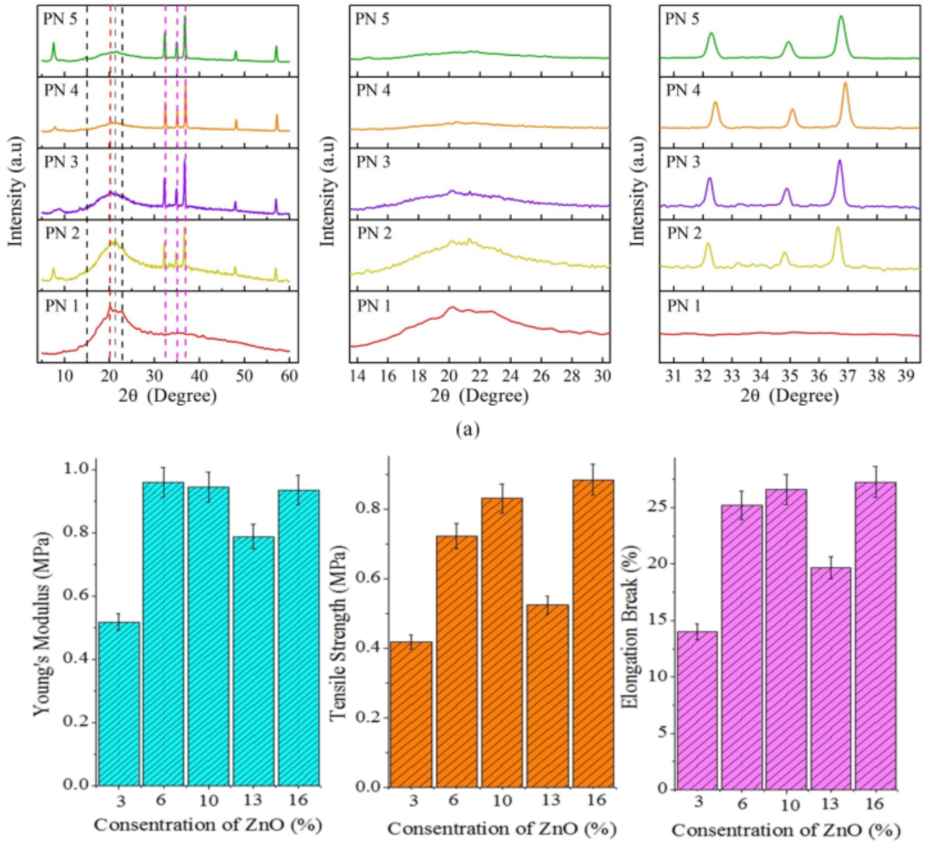
**Fig. 4.** (a) The Fourier transform infra-red (FTIR) spectra (1 for chitosan/cellulose and 5%wt ZnO (CCZ) [23], 2 for starch/chitosan and 3 % Pineapple leaf microfibers (PLM) [9], and 3 for chitosan and 5%wt of ZnO composite [25]) (b) surface image from scanning electron microscope (SEM) of PN 1 for 1, PN 3 for 2, and PN 5 for 3, and chitosan-0.5 % ZnO for 4 from Ref. [26] for comparison. (c and d) schematic illustration of three- and two-dimensional view of (c) composite starch and chitosan (d) for composite starch and chitosan with additional nanocrystal ZnO and Pineapple leaf fiber (PALF). (e) schematic bonding between starch and chitosan from via OH (upper) and for additional ZnO and PALF also from OH bonding and some of the freely molecule $N^{-2}$, OH, $H_2O$ (bottom). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

shows surface image from scanning electron microscope (SEM) of PN 1 for 1, PN 3 for 2, and PN 5 for 3. The surface image clearly shows that the ZnO formation, for PN 1 shows inhomogeneous particle distribution, for PN 3 shows the breaking of PLF and some of particle attached the PLF probably from ZnO. For PN 5 as the highest amount of ZnO shows the particle dominant at the surface that probably hinder the PLF at the surface film. The SEM surface image in this study shows similar image for CCZ from Ref. [23], chitosan-0.5 % ZnO from Ref. [26], and we have included in Fig. 4 indicated by label 4 (b) from Ref. [26] for comparison. The OH bond contributed to intramolecular and intermolecular hydrogen bonds which play an important role in bonding with PALF and ZnO by crosslinking $Zn^{2+}$ ions with PLF through electrostatic interactions and generation of ZnO nanoparticles with PLF as a capping agent [48]. The intermolecular hydrogen bonds were weakened, and new hydrogen bonds were formed between starch and chitosan as shown in Fig. 3 (c) that make easier movement molecular chain in form of translation and rotational. When PALF and ZnO was introduced, the new hydrogen bond from the starch was formed and bonding with PALF and ZnO (Fig. 4 (d)). Some of the molecules break the bonding such as ZnO forming $Zn^{2+}$ and $O^{2-}$, and chitosan and starch also produced freely molecule $N^{-2}$, OH, $H_2O$ as shown in Fig. 4 (e).

### 3.4. XRD (X-ray diffraction) spectra

Fig. 5 (a) shows the results of characterization of variations in PN samples with the main ingredients of starch, chitosan, PALF, and ZnO. Based on the research results, the diffraction peak of cassava starch is in the range of $2\theta = 18°$-$20°$ (red dash line) and chitosan is in the range of $2\theta = 21°$- $22°$ (grey dash line) which is a semi-amorphous structure. Sample PN 1 in the range $2\theta = 30°$-$40°$ (violet dash line) which is the crystal structure of ZnO shows a very small.

diffraction peak intensity as well as for the diffraction peak of PALF. Based on the results of research by (Mutmainna, et al., 2019) showed the peak of the combination of chitosan and PALF was at $2\theta = 34°$ and for the diffraction peak of PALF itself was at $2\theta = 48°$ [9]. Another case obtained in the study of (Natalia J et al., 2018) stated that the diffraction peaks of PALF were at $2\theta = 15°$ and $23°$ [49]. Similar results were also obtained by (Balakrishnan et al., 2017) at $2\theta = 15°$ and $23°$ [8] and



(a)

(b)

**Fig. 5.** (a) XRD full spectra (left), enlarging diffraction peak at 18-24° (middle), and at 31-39° (right) and (b) Young's modulus (left), Tensile Strength (middle), and elongation at break (right) of PN samples for various concentration of ZnO (PN 1-PN 5).

Amalia et. Al [50] at $2\theta = 15.25°$ and 20.46°.

For sample PN 1 there are three diffraction peaks with the highest intensity, namely at 2 $2\theta = 20.16°$, 21.18°, and 22.60°, where these peaks are a combination of starch and chitosan. As well as visible diffraction peaks from PALF, namely at $2\theta = 15.74°$ and 23.56° (black dash line), but the diffraction peak for ZnO is not visible due to the small intensity of ZnO. There is also a small diffraction peak in the range of $2\theta = 28°$-30° for samples PN 1–PN 5 which is the diffraction peak of chitosan, where the greater the concentration of ZnO, the lower the intensity of chitosan. The PN 2 sample contained three ZnO diffraction peaks with the highest intensity at $2\theta = 32.15°$, 34.79°, and 36.62° and in the range, $2\theta = 18°$-22° the intensity of the diffraction peaks decreased. Fig. 4(a) also shows that the addition of ZnO concentration increases the peak intensity at $2\theta = 31°$-36°, which can be seen in samples PN 2–PN 5. In sample PN 3, the intensity of the diffraction peak in the range of 2θ for starch/chitosan decreases while the intensity of the diffraction peak decreases. 2θ for ZnO increased at $2\theta = 32.20°$, 34.85°, and 36.68°. Similar results were also obtained in the PN 4 sample, the intensity of the 2θ diffraction peak for starch/chitosan decreased while the intensity of the 2θ diffraction peak for ZnO increased at 32.40°, 35.05°, and 36.88° and in the PN 5 sample the peak intensity 2θ diffraction for ZnO at 32.26°, 34.92° and 36.74°. These results indicate that the crystallinity of the mixture of starch/chitosan/PALF decreased with the addition of ZnO concentration in the bioplastic samples. The crystallite size from the diffraction peaks of ZnO are 0.33 nm for PN 1, 0.33 nm for PN 2, 0.32 nm for PN 3, 0.34 nm for PN4, and 0.33 nm for PN 5 which similar reported in Ref. [25].

### 3.5. *Mechanical properties*

The mechanical properties in Fig. 5 (b) shows Young modulus, tensile strength, and elongation at break for bioplastics with additional PALF of PN samples. Based on the results obtained, as ZnO increased in the cassava starch/chitosan/PALF matrix, the elongation at break values increased from PN 1 to PN 3, respectively 15.516, 21.669, and 24.897 MPa. These phenomena for elongation at break are same for Young modulus and tensile strength.

The increase in elongation at break indicates that a good interfacial bond between ZnO and the starch matrix occurs. This interfacial bond is an important aspect that affects to the mechanical properties [51]. The strong interaction between the starch matrix and ZnO nanofillers was due to the homogeneous dispersion of ZnO in the starch matrix, thus the mechanical properties increase. However, at PN 4 the at break value decreased to 23.634 MPa. This could be due to the agglomeration of ZnO, inhomogeneous ZnO dispersion causes a weakening of the starch PALF-ZnO matrix interaction [52] and at PN 5 the elongation at break value increased to 26.516 MPa. Based on the results of mechanical properties indicated that the application of PALF helping the performance of ZnO as a reinforcement. The same thing was also expressed by the research of Gloria et al. [14] that, pineapple leaf fiber significantly increased the elongation at break of the matrix composite. Thus, based on the previous explanation, bioplastics made from cassava starch have a lower mechanical properties value than starch/chitosan bioplastics and pineapple leaf fiber. The addition of ZnO also significantly affects for the improvement of the mechanical properties of bioplastics.

### *3.6. Packaging test*

Bacterial or fungal growth is the two main problems in bakery products. As a solution, ZnO-based bio-composite for bioplastic due to the antimicrobial which is expected to increase the shelf life of sliced white bread. Fig. 6 shows the visual results of the shelf life of microbes in white bread wrapped without and using bioplastic.

Fig. 6 shows the visual results of the variation of PN 1-PN 5 samples. It clearly shows that after 10 days, the control sample (sample unwrapped in bioplastic) was overgrown with microbes which were characterized by blackening of the bread and the texture of the bread was harder (staling) than usual or showed high amylopectin retrogradation. Compared to the PN sample, the sample was overgrown by microbes after 30 days, this was because the type of starch affected the rate of retrogradation [3].

It was found that, when the concentration of ZnO increases, microbes are difficult to grow. In PN 1-PN 3 samples, microbes began to grow on day 30. When compared to PN 1-PN 3, microbial growth was faster in samples PN 1 and continued to decrease in samples PN 2 to PN 3. As for samples PN 4–5 has not been overgrown by microbes, but the texture of the whole bread tends to dry out. This shows that the antimicrobial activity increases rapidly with the presence of ZnO in the polymer matrix. Electrostatic binding of $Zn^{+2}$ ions to the cell surface of microorganisms, causes changes in cell membrane permeability, interactions with respiratory enzymes and their inactivation, reactions with cell components, and depletion of cytoplasmic contents. Thus, microbes will be difficult to live [53].

The Ref. [23] reported that the chitosan high hydrophilicity due to the hydrogen bonding formation between $NH_2$ group of chitosan with OH group of PALF which increase hydrophilicity [54]. Addition of nano ZnO particles imparted significant hydrophobicity to the composite as reported by [3], for the concentration ZnO increase (PN5), the hydrophobicity increased due to the dispersion of nano ZnO in starch/chitosan matrix [28]. The nano ZnO have high barrier ability for water due to the generation reactive species such as $OH^-$, $H_2O_2$ and $O_2^{-2}$. These reactive species come from the defect of ZnO were activated by the photon from the UV or visible light. These photons created ($e^-$) and ($h^+$), the ($h^+$) break the bond of $H_2O$ molecules resulting $OH^-$ and $H^+$, the reaction mechanism as follows [55–57]:

$$ZnO + h\nu \rightarrow ZnO\ (e^-) + ZnO\ (h^+) \tag{3}$$

at the valence band of ZnO:

$$ZnO\ (h^+) + H_2O \rightarrow ZnO + H^+ + OH^{-*} \tag{4}$$

at the conduction band of ZnO:

$$ZnO\ (e^-) + O_2 \rightarrow ZnO + O_2^{*-} \tag{5}$$

$$O_2^{*-} + H^+ \rightarrow OOH^* \tag{6}$$

$$O_2^{*-} + H_2O \rightarrow OOH^* + OH^- \tag{7}$$

$$OOH^* + H^+ + ZnO\ (e^-)) \rightarrow H_2O_2 + ZnO \tag{8}$$

$$OOH^* + H_2O_2 + ZnO\ (e^-) \rightarrow H_2O_2 + OH^- + ZnO \tag{9}$$

The negatively charged reactive particles attach only in outer surface of bacteria and cannot penetrate the cell membrane. The molecule which can penetrate the cell membrane and kill the bacteria is $H_2O_2$ as a result of chemical reaction process start from the defect of ZnO activated by the photon from the UV or visible light and finally as shown in Eqs. (8) and (9) [7].

## 4. Conclusion

For 16 % of ZnO is the maximum mechanical properties with new structure was formed as the contribution from each base material. The elongation at break values increased from PN 1 to PN 3, respectively 15.516, 21.669, and 24.897 MPa. For PN 4 the elongation at break decreased to 23.634 MPa due to the agglomeration of ZnO causes a weakening of the starch PALF-ZnO matrix interaction and at PN 5 increased to 26.516 MPa. The bioplastics PALF/ZnO-based was successfully degraded easily in soil for 21 days and only 18 days in the seawater. The results of packaging tests, improvement in antimicrobial properties obtained for coatings contains 10,13, and 16 % ZnO NPs with no fungal growth for 30 days. The bioplastics PALF ZnO-based have high potential to be used for food packaging with faster biodegradation performance and inhibit bacterial growth.

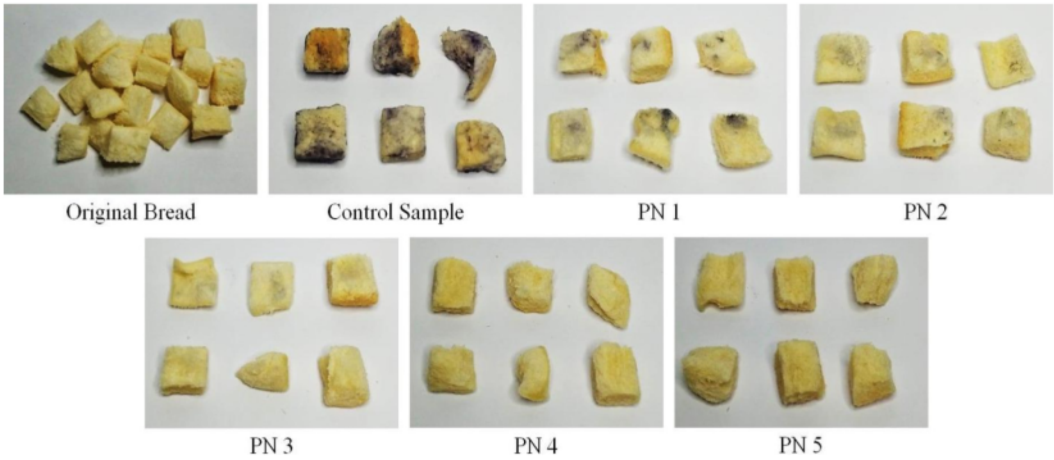

Original Bread
Control Sample
PN 1
PN 2
PN 3
PN 4
PN 5

**Fig. 6.** Visual photograph results of bioplastics packaging test on bread. First rows from the left are original bread, control sample without packaging for 10 days, PN 1-PN 2 for 30 days, and PN 3- PN 5 continue to the second rows for 30 days.

## CRediT authorship contribution statement

**Bidayatul Armynah:** Investigation, Methodology, Writing – original draft, Writing – review & editing. **Rahma Anugrahwidya:** Investigation, Methodology, Data curation, Resources, Writing – original draft. **Dahlang Tahir:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Resources, Supervision.

## Declaration of competing interest

The author declare that they have no known competing financial or personal relationship that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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## References

[1] A. Shafqat, T. Arifa, M. Adeel, B Amtul .T., Y. Abdullah, P. Arivalagan, A review on environmental significance carbon foot prints of starch based bio-plastic: a substitute of conventional plastics, Biocatal. Agric. Biotechnol. 27 (2018), 101540.

[2] S Bhatia .K., V Sachin .O., M. Jong, G. Ranjit, K Yong .C., K Ravi .B., P. Arivalagan, K. Vinod, J Rajesh .B., J Jeong .Y., Y. Kwon, H Yung .Y., Biowaste-to-bioplastic (Polyhydroxyalkanoates): conversion technologies, strategies, challenges, and perspective, Bioresour. Technol. 326 (2021), 124733.

[3] N. Noshirvani, B. Ghanbarzadeh, R. Rezaei Mokarram, M. Hashemi, Novel active packaging based on Carboxymethyl cellulose-chitosan-ZnO NPs nanocomposite for increasing the shelf life of bread, Food Packag. Shelf Life 11 (2018) 106–114.

[4] R. Zhang, X. Wang, M. Cheng, Preparation and characterization of potato starch film with various size of Nano-$SiO_2$, Polym. J. 10 (10) (2018) 1172.

[5] Harunsyah., M. Yunus., F. Reza, Mechanical properties of bio-plastics cassava starch film with zinc oxide Nanofiller as reinforcement, in: IOP Conf. Series: Mater. Sci. Eng 210, 2017 (012015).

[6] F Titani .R., Haryanto, The effect of Nano-zinc oxide and titanium dioxide on the properties of bioplastic, Int. J. Adv. Sci. Technol. 6 (2) (2018) 2321–8991.

[7] A Abdullah .H,.D., D Oceu .P., K Anti .F., C Rossy .N., H. Syamsul, F Rahmat .S., K. Myrtha, S. Rahmat, Harnessing the excellent mechanical, barrier and antimicrobial properties of zinc oxide (ZnO) to improve the performance of starch-based bioplastic, Polym. Plast. Technol. Mater. 59 (12) (2020) 1259–1267.

[8] P. Balakrishnana, M Sreekala .S., K. Matjaz, H. Miroslav, T. Sabu, Morphology, transport characteristics and viscoelastic polymer Chainconfinement in nanocomposites based on thermoplastic potato Starchand cellulose nanofibers from pineapple leaf, Carbohydr. Polym. 169 (2017) 176–188.

[9] I. Mutmainna, D. Tahir, L. Paulus, I. Sultan, Synthesis composite starch-chitosan as biodegradable plastics for food packaging, in: IOP Conf. Series: Journal of Physics: Conf. Series 1317, 2019 (012053).

[10] P. Sahu, M Gupta .K., Sisal (Agave Sisalana) Fiber and its polymer-based composites: a review on current developments, Reinf. Plast. Compos. 36 (24) (2017) 1759–1780.

[11] A Almusawi .M., T Hussein .S., M Shallal .A., Effect of temperature and sisal Fiber content on the properties of plaster of Paris, Int. J. Eng. Technol. 7 (4.20) (2018) 205–208.

[12] S. Edi, Jamaluddina., W. Sentot, A. Irwan, A. Mochamad, H.S. Nasmi, F. Ahmad, Characterization and properties of cellulose microfibers from water hyacinth filled sago starch biocomposites, Int. J. Biol. Macromol. 137 (2019) 119–125.

[13] Y. Libo, C. Nawawi, Y. Xiaowen, Improving the mechanical properties of natural fibre fabric reinforced epoxy composites by alkali treatment, J. Reinf. Plast. Compos. 31 (6) (2012) 425–437.

[14] G Gloria .O., G.R. Altoé, M.A. Gomes, C.M.F. Vieira, M.C.A. Teles, F.M. Margem, Izod toughness behavior of continuous PALF reinforced polyester matrix composites, in: Characterization of Minerals, Metals and Materials, Ham, Switzerland, Springer Nature, 2017, pp. 41–47.

[15] T Gutiérrez .J., P.G. Seligra, C.M. Jaramillo, L. Famá, S. Goyanes, Effect of filler properties on the antioxidant response of thermoplastic starch composites, in: Handbook of Composites from Renewable Materials, Structure and Chemistry 1, 2017, pp. 337–370.

[16] R. Anugrahwidya, Y. Nurfina, D. Tahir, Optical and structural investigation of synthesis ZnO/ag nanoparticles prepared by laser ablation in liquid, Mater. Sci. Semicond. Process. 105 (2020), 104712.

[17] M Arrieta .P., L. Peponi, D. López, J. López, J.M. Kenny, An overview of nanoparticles role in the improvement of barrier properties of bio-plastics for food packaging applications, in: Food Packag, Academic Press, 2017, pp. 391–424.

[18] R. Anugrahwidya, B. Armynah, D. Tahir, Composites bioplastic film for various concentration of zinc oxide (ZnO) nanocrystals towards physical properties for high biodegradability in soil and seawater, J. Polym. Environ. 30 (2022) 2589–2601, https://doi.org/10.1007/s10924-021-02363-4.

[19] E Nanang .W., S. Heru, Analysis of biodegradation of bioplastic made of cassava starch, J. Mech. Eng. Sci. 1 (1) (2017) 24–31.

[20] B. Montero, M. Rico, L Rodríguez .S., L. Barral, R. Bouza, Effect of nanocellulose as a filler on biodegradable thermoplastic starch films from tuber, cereal and legume, Carbohydr. Polym. 157 (2017) 1094–1104.

[21] A Nafchi .M., R. Assiri, S. Sheibani, F. Ariffin, A.A. Karim, Preparation and characterization of bionanocomposite films filled with nanorod-rich zinc oxide, Carbohydr. Polym. 96 (2013) 233–239.

[22] M Hoque .B., M.S. Hossain, M.N. Abdul, B. Solaiman, A.K. Ruhul, Fabrication and characterization of pineapple fiber-reinforced polypropylene based composites, in: Nano Hybrids and Composites vol. 21, 2018, pp. 31–34.

[23] M.P. Indumathi, K. Saral Sarojini, G.R. Rajarajeswari, Antimicrobial and biodegradable chitosan/cellulose acetate phthalate/ZnO nano composite films with optimal oxygen permeability and hydrophobicity for extending the shelf life of black grape fruits, Int. J. Biol. Macromol. 132 (2019) 1112–1120, https://doi.org/10.1016/j.ijbiomac.2019.03.171.

[24] M.S.A. Darwish, M.H. Mostafa, L.I. Hussein, A.H. Abdaleem, M.A. Elsawy, Preparation, characterization, mechanical and biodegradation behavior of polypropylene - chitosan/ZnO nanocomposite, Polym. Plast. Technol. Mater. 1–11 (2021), https://doi.org/10.1080/25740881.2021.1924200.

[25] S. Ummartyotin, C. Pechyen, Physico-chemical properties of ZnO and chitosan composite for packaging material, J. Biobased Mater. Bioenergy 11 (2017) 183–192, https://doi.org/10.1166/jbmb.2017.1661.

[26] S. Sanuja, A. Agalya, M.J. Umapathy, Synthesis and characterization of zinc oxide–neem oil–chitosan bionanocomposite for food packaging application, Int. J. Biol. Macromol. 74 (2015) 76–84, https://doi.org/10.1016/j.ijbiomac.2014.11.036.

[27] L.-H. Li, J.-C. Deng, H.-R. Deng, Z.-L. Liu, L. Xin, Synthesis and characterization of chitosan/ZnO nanoparticle composite membranes, Carbohydr. Res. 345 (8) (2010) 994–998, https://doi.org/10.1016/j.carres.2010.03.019.

[28] P. Mujeeb Rahman, V.M. Abdul Mujeeb, K. Muraleedharan, S.K. Thomas, Chitosan/nano ZnO composite films: enhanced mechanical, antimicrobial and dielectric properties, Arab. J. Chem. 11 (1) (2018) 120–127, https://doi.org/10.1016/j.arabjc.2016.09.008.

[29] D.S. Vicentini, A. Smania, M.C.M. Laranjeira, Chitosan/poly (vinyl alcohol) films containing ZnO nanoparticles and plasticizers, Mater. Sci. Eng. C 30 (4) (2010) 503–508, https://doi.org/10.1016/j.msec.2009.01.026.

[30] L. Suryanegara, W. Fatriasari, D. Zulfiana, S.H. Anita, N. Masruchin, S. Gutari, T. Kemala, Novel antimicrobial bioplastic based on PLA-chitosan by addition of TiO2 and ZnO, J. Environ. Health Sci. Eng. 19 (1) (2021) 415–425, https://doi.org/10.1007/s40201-021-00614-z.

[31] L. Sapei, K.S. Padmawijaya, O. Sijayanti, P.J. Wardhana, Study of the influence of ZnO addition on the properties of chitosan-banana starch bioplastics, in: IOP Conference Series: Materials Science and Engineering 223, 2017, https://doi.org/10.1088/1757-899x/223/1/012044 (012044).

[32] X. Hu, X. Jia, C. Zhi, Z. Jin, M. Miao, Improving the properties of starch-based antimicrobial composite films using ZnO-chitosan nanoparticles, Carbohydr. Polym. (2019), https://doi.org/10.1016/j.carbpol.2019.01.043.

[33] X. Li, H. Li, X. Wang, D. Xu, T. You, Y. Wu, F. Xu, Facile in situ fabrication of ZnO-embedded cellulose nanocomposite films with antibacterial properties and enhanced mechanical strength via hydrogen bonding interactions, Int. J. Biol. Macromol. 183 (2021) 760–771, https://doi.org/10.1016/j.ijbiomac.2021.04.175.

[34] F. Yu, X. Fei, Y. He, H. Li, Poly(lactic acid)-based composite film reinforced with acetylated cellulose nanocrystals and ZnO nanoparticles for active food packaging, Int. J. Biol. Macromol. 186 (2021) 770–779, https://doi.org/10.1016/j.ijbiomac.2021.07.097.

[35] F. Fu, J. Gu, R. Zhang, X. Xu, X. Yu, L. Liu, J. Yao, Three-dimensional cellulose based silver-functionalized ZnO nanocomposite with controlled geometry: synthesis, characterization and properties, J. Colloid Interface Sci. 530 (2018) 433–443, https://doi.org/10.1016/j.jcis.2018.07.009.

[36] K. Dharmalingam, R. Anandalakshmi, Functionalization of cellulose-based nanocomposite hydrogel films with zinc oxide complex and grapefruit seed extract for potential applications in treating chronic wounds, Polymer 122620 (2020), https://doi.org/10.1016/j.polymer.2020.122620.

[37] E. Lizundia, A. Urruchi, J.L. Vilas, L.M. León, Increased functional properties and thermal stability of flexible cellulose nanocrystal/ZnO films, Carbohydr. Polym. 136 (2016) 250–258, https://doi.org/10.1016/j.carbpol.2015.09.041.

[38] Y. Xie, Y. Pan, P. Cai, Cellulose-based antimicrobial films incroporated with ZnO nanopillars on surface as biodegradable and antimicrobial packaging, Food Chem. 368 (2022), 130784, https://doi.org/10.1016/j.foodchem.2021.13078.

[39] H. Abral, N. Fajri, M. Mahardika, D. Handayani, E. Sugiarti, H.-J. Kim, A simple strategy in enhancing moisture and thermal resistance and tensile properties of disintegrated bacterial cellulose nanopaper, J. Mater. Res. Technol. 9 (4) (2020) 8754–8765, https://doi.org/10.1016/j.jmrt.2020.06.023.

[40] Y. Li, H. Sun, Y. Zhang, M. Xu, S.Q. Shi, The three-dimensional heterostructure synthesis of ZnO/cellulosic fibers and its application for rubber composites, Compos. Sci. Technol. (2019), https://doi.org/10.1016/j.compscitech.2019.04.012.

[41] M. Wu, Z. Zhou, J. Yang, M. Zhang, F. Cai, P. Lu, ZnO nanoparticles stabilized oregano essential oil Pickering emulsion for functional cellulose nanofibrils packaging films with antimicrobial and antioxidant activity, Int. J. Biol. Macromol. 190 (2021) 433–440, https://doi.org/10.1016/j.ijbiomac.2021.08.210.

[42] X. Li, Z. Ren, R. Wang, L. Liu, J. Zhang, F. Ma, X. Liu, Characterization and antibacterial activity of edible films based on carboxymethyl cellulose, Dioscorea opposita mucilage, glycerol and ZnO nanoparticles, Food Chem. 349 (2021), 129208, https://doi.org/10.1016/j.foodchem.2021.129208.

[43] R. Anugrahwidya, A. Bidayatul, D. Tahir, Bio-plastics starch-based with additional Fiber and nanoparticle: characteristics and biodegradation performance: a review, J. Environ. Sci. 29 (11) (2021) 3459–3476.

[44] J. Jamiluddin, P Januar .S., N.O. Ahmed, H.M.H. Mohammad, T. Cionita, S. S. Mohd, Experimental investigation on performance of short pineapple leaf Fiber reinforced tapioca biopolymer composites, BioResources. 13 (3) (2018) 6341–6355.

[45] M. Mittal, C. Rajiv, Biodegradability and mechanical properties of pineapple leaf/coir Fiber reinforced hybrid epoxy composite, Mater. Res. Express 6 (2019), 045301.

[46] A. Jayakumar, K Heera .V., T.S. Sumi, M. Joseph, S. Mathew, G. Praveen, I.C. Nair, E.K. Radhakrishnan, Starch-PVA composite films with zinc-oxide nanoparticles and phytochemicals as intelligent pH sensing wraps for food packaging application, Int. J. Biol. Macromol. 136 (2019) 395–403.

[47] X. Zhang, F. Wang, L. Keer, Influence of surface modification on the microstructure and Thermo-mechanical properties of bamboo fibers, Materials 8 (10) (2015) 6597–6608, https://doi.org/10.3390/ma8105327.

[48] Q. Bai, L. Yangchao, Preparation and characterization of Carboxymethyl cellulose capped zinc oxide nanoparticles: a proof-of-concept study, Food Chem. 389 (2022), 133001.

[49] J. Natalia, M Juan .V., M.C.C. Edith, R.O. Adriana, Improvement of mechanical properties of pineapple leaf fibers by mercerization process, Fibers Polym. 19 (12) (2018) 2604–2611.

[50] B. Amalia, C. Imawana, A. Listyarini, Effect of Nanofibril cellulose isolated from pineapple leaf on the mechanical properties of chitosan film, AIP Conf. Proc. 2023 (2018), 020034.

[51] D. Tahir, S Oh .K., H.J. Kang, S. Tougaard, Composition dependence of dielectric and optical properties of Hf-Zr-silicate thin films grown on Si(100) by atomic layer deposition, Thin Solid Films 616 (2016) 425–430.

[52] S. Gamero, R Jiménez .M., A. Romero, C. Bengoechea, A. Guerrero, Reinforcement of soy protein-based bioplastics through addition of lignocellulose and injection molding processing conditions, J. Polym. Environ. 27 (6) (2019) 1285–1293.

[53] T. Bhuyan, K. Mishra, M. Khanuja, R. Prasad, A. Varma, Biosynthesis of zinc oxide nanoparticles from Azadirachta Indica for antibacterial and photocatalytic applications, Mater. Sci. Semicond. Process. 32 (2015) 55–61.

[54] F.M. Pelissari, M.V. Grossmann, F. Yasmashita, E.A.G. Pineda, Antimicrobial, mechanical and barrier properties of cassava starch-chitosan films incorporated with oregano essential oil, J. Agric. Food Chem. 57 (2009) 7499–7504.

[55] K. Qi, B. Cheng, J. Yu, W. Ho, Review on the improvement of the photocatalytic and antibacterial activities of ZnO, J. Alloys Compd. 727 (2017) 792–820, https://doi.org/10.1016/j.jallcom.2017.08.142.

[56] S. Baruah .K., S. Pal, J. Dutta, Nanostructured zinc oxide for water treatment, Nanosci. Nanotechnol. Asia 2 (2) (2012) 90–102.

[57] T.S. Tofa, E. Plaza, J. Dutta, Degradation of microplastic residuals in water by visible light Photocatalysis, Environ. Eng. Sustain. Infrastruct. (2018) 11–13.

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| | Mufti Hatur Rahmi, Dahlang Tahir. "The Correlation between Structural and Optical Properties of Zinc Hydroxide Nanoparticle in Supports Photocatalytic Performance", Optical Materials, 2021<br>Publication | |
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---

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---

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---

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---

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| | | |
|---|---|---|
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| | | |
|---|---|---|
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---

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---

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---

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---

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40 Wenkai Hu, Zhiming Zou, Heping Li, Ziang Zhang, Jingling Yu, Qun Tang. "Fabrication of highly transparent and multifunctional polyvinyl alcohol/starch based nanocomposite films using zinc oxide nanoparticles as compatibilizers", International Journal of Biological Macromolecules, 2022
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|---|---|---|
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| | | |
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| | | |
|---|---|---|
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