RESEARCH PROGRESS IN MECHANICAL AND
MANUFACTURING ENGINEERING
e-ISSN: 2773-4765

RPMME

Vol. 6 No. 1 (2025) 131-137
https://publisher.uthm.edu.my/periodicals/index.php/rpmme

Preparation and Characterization of Potato Starch-Based Bioplastic for Food Packaging Applications

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DOI: https://doi.org/10.30880/rpmme.2025.06.01.013

Article Info

Received: 01 November 2024
Accepted: 01 July 2025
Available online: 31 July 2025

Keywords

Bioplastic, Chitosan, Crystallinity, Potato Starch, Surface Morphology, Zinc Oxide

Abstract

The hazardous impact of fossil fuel-based plastics on the environment and society has drawn attention toward alternative sources. Bioplastics are a type of renewable resource that can be degraded by microorganisms and prevent chemical leaching. So, the starch-based bioplastics properties became an important factor to replace fossil fuel-based plastics. This study aims to characterize the addition of both Zinc Oxide and Chitosan to potato starch bioplastics films. Bioplastic potato starch-based films were prepared by using the casting method. The concentration of zinc oxide in the bioplastics samples varies from 1.0 %, 2.0%, and 3.0% (w/w) by weight of starch. Also, the chitosan content in the bioplastic varies from 4.0%, 6.0%, and 8.0% (w/w) by weight of starch. The result of the SEM samples shows that the surface becomes smoother as the zinc oxide and chitosan content is higher. Sample 3 shows the lowest crystallinity value, which is 74%, determined by X-ray diffraction analysis (XRD), indicating a high degree of degradation. Samples 7 and 9 had 79% crystallinity value, which is the highest, which is represented by better mechanical properties and longer shelf life. The FTIR data show the IR spectra presented the existence of O-H groups, C-H groups, C=C groups and C-O-C ester groups. The change of group functions was analysed, and no changes occurred. So, there was only physical interaction in the bioplastic samples.

1. Introduction

In the 21st century, the use and application of petrochemical-based plastics are increasing day by day due to the growing population. Consequently, there is an increase in environmental pollution due to the non-decomposition of petrochemical-based plastics [1]. In food packaging industries, plastic is one of the most applicated materials due to its chemical-resistance, thermal resistance, and corrosion resistance. The excellent performance of petrochemical-based plastic such as lightweight, cheap, and waterproof make it the most used in food packaging industry [2]. Petrochemical-based plastics are known as one of the artificial polymer materials. It is known as non-decomposition and non-biodegradable because they defy microbial degradation. The

Petrochemical-based plastics often take a long time, up to hundreds or even thousands of years, to break the carbon chain, then end up in landfills and damage the environment.

The non-biodegradable plastics cause them to be discarded. The plastics discarded ultimately cause plastic pollution if the plastics are not well managed. According to [1], the single-use plastics (disposable) escape from the collection and recycling system is approximately 10 MT/year, which is roughly 32% or even higher. This includes the plastics that are escaping into the water system, ocean, and marine environment. The plastics pollution of the ocean and marine system is macroplastics (large plastics waste such as straws and plastic bags) and microplastics (smaller than 5mm in size). Microplastic pollution is not only causing environmental problems, but it also triggers human health problems. Inhalation and ingestion of microplastics accumulate in the human body, which can trigger the immune system or cause local toxicity. Not only that, microplastics accumulate in the bodies of organisms, causing bioaccumulation in ecosystems. So, fully biodegradable plastic is necessary for decreasing the environmental impact [3].

Bioplastics is being introduced as one of the plastics with high biodegradability. Therefore, replacement of single-use plastics (disposable) with bioplastics requires the bioplastics to have similar properties, including physical and mechanical [4]. Bioplastics are an alternative way to reduce and replace petrochemical-based plastics because bioplastics are environmentally friendly, renewable, and biodegradable in a short time. Bioplastics made from renewable polymers and biodegradable, including a variety of natural starch, natural oils, protein, and cellulose, have attracted considerable attention [5]. The main advantage of biodegradable plastic is they are not necessary to recycle because they have a shelf life. This is because they involve a biodegradation process. Organic molecules in the environment are broken down into simpler compounds by breaking bonds through hydrolysis or by bacteria, fungi, yeast, and their enzymes in the polymer leading polymer erosion.

Biodegradable plastics are generally classified into two groups: (1) natural (e.g. starch, cellulose, gelatin etc) and (2) synthetic (e.g. polyester). Starch is a type of cheap and biodegradable biopolymer which can be easily obtained from plant resources. It is a renewable resource with easily modifiable biopolymer which made starch one of the best raw materials to produce starch-based bioplastic. Starch based plastics offer lower cost materials than some other types of biodegradable polymers such as synthetic polyesters and polylacitc acid (PLA) because of relatively cheap agricultural feedstock and simpler manufacturing process which becomes the market drivers of starch-based polymers [6]. In addition, starch-based plastics are more environmentally friendly than synthetic bioplastics. Various types of bags such as shopping bags, garage bags, and compost bags are made from starch-based polymers. It has also found use in rigid packaging applications including containers and trays for packaging fresh food and convenience food. Also, bioplastics meet the benchmark performance of petrochemical-based plastic is necessary in the food packaging industry. This bioplastic must require characteristics such as lightweight, waterproof, and thermal-resistance to replace the petrochemical-based plastics in food packaging industries. Properties of certain bioplastics nowadays are facing problems like thermal instability, difficult heat saleability, brittleness, low melt strength, high water vapor and oxygen permeability which have limited their use as film in food packaging applications [7].

The bioplastics in this study focused on starch-based bioplastics. This is because many reports have successfully derived bioplastics from various types of starch and have been documented. For cassava starch bioplastics, the foam tray produced had almost similar mechanical properties to the polystyrene foam [8] and the mechanical properties are influenced by the concentration of glycerol and starch [9]. For corn starch bioplastics, the bioplastics by using the casting method will normally have a lower average thickness due to the loss of water content at the surface [10], and the presence of taro starch will improve the mechanical properties of the corn starch bioplastics [11]. The amount of research on the mechanical properties of potato starch is the least compared to other types of starch due to its properties and performance [12]. But from the aspect of availability, potato starch is the world's third highest produced starch [13]. So, potato starch in casting bioplastics is still under discovery and lack of research.

In the present study, potato-starch based bioplastics is cast by using potato starch, glycerol, and acetic acid with the addition of ZnO and chitosan as an additive. ZnO and Chitosan are used to enhance the performance of potato-starch based bioplastics. ZnO is used to increase the mechanical properties and shelf life while chitosan is used to increase the biodegradability. The reinforcing effect of various concentrations of ZnO and chitosan on the characterization of bioplastics is investigated by using FTIR, SEM and XRD. FTIR provides information about the group function of bioplastics and the mechanical properties were determined as bioplastics have a similar group function to the previous study. Then, the SEM determined the surface morphology of the bioplastic samples. The XRD analyzes the crystallinity of the bioplastic samples, and the biodegradability was estimated.

2. Materials and methods

2.1 Materials

Bestari potato starch was provided by Synechem Food Processing Ind. Sdn. Bhd. Acetic acid and glycerol with food grade. 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), sulfuric acid (H_2SO_4) solution p.a. ≥25 %, ZnO powder with maximum particle size 45-micron. All aqueous solution using deionized water.

2.2 Preparation of samples

The potato starch is prepared in different sets with the same mass. The sample's thickness is at 0.75 ± 0.05mm, which is lower than 1mm. Samples with the potato starch were named with numerical 1-9 which stands for the sample 1-9. Firstly, the chitosan is prepared in different masses, each is mixed with 25% sulphuric acid, which is double the mass of chitosan, under stirring (400rpm) and heating (110°C). For example, 0.67g of chitosan mixed with 1.34g of sulphuric acid. Then, filmogenic solution is prepared by dissolving the potato starch in acetic acid, deionized water and glycerol. Then, the mixture of chitosan and sulphuric acid is mixed with the filmogenic solution under stirring until the solution temperature reaches between 60 and 63 63^{o}C. The filmogenic solution is stirred with a magnetic stirrer at 800 rpm until the internal temperature is reached. Then, the filmogenic solution is poured into the mould prepared for cooling and shaping. Table 1 shows the composition of the samples.

Table 1 *The composition of various bioplastic samples.*

Sample	Potato starch (g)	Glycerol (ml)	Acetic acid (ml)	Deionized water (ml)	ZnO (g)	Chitosan (g)
1	16	9	5	70	0.17	0.64
2	16	9	5	70	0.17	0.96
3	16	9	5	70	0.17	1.28
4	16	9	5	70	0.33	0.64
5	16	9	5	70	0.33	0.96
6	16	9	5	70	0.33	1.28
7	16	9	5	70	0.50	0.64
8	16	9	5	70	0.50	0.96
9	16	9	5	70	0.50	1.28

2.3 Bioplastic film sample test preparations

The Fourier transform infrared (FTIR) (Perkin Elmer) was used to analyze the bonding and group function of the bioplastic samples. The bioplastics samples are cut into 1cm x 1cm for the FTIR testing. X-ray diffraction (XRD) (Bruker D8 Advance) is used to examine the crystallinity of bioplastic samples. The instrument is set with the degree measurements set from $2\theta = 10.01^\circ$ until 89.9° with beam wavelengths of $(\lambda = 1.54Å)$ and ($\lambda = 1.39Å$) at 25℃ [3]. The operating current was set at 35mA. The potential difference is set at 40kV. The vivid peaks and valleys of the XRD diagram of each sample are determined. The crystallinity value (Cr) is calculated by using Equation (1)

$$Cr = 100 \frac{I_{crystallinity}}{I_{amorphous} + I_{crystallinity}} \% \qquad (1)$$

The Scanning Electron Microscopy (SEM) (HITACHI SU1510) pictures were taken employing an electrical potential for accelerating operating at 10kV, and picture magnifications were set at 500x. The working distance (WD) and the spot calibration are set at 10.8mm and 5.5mm, respectively. The pictures taken using SEM are being analyzed for surface morphology. The surface morphology is the key to determining the mechanical properties of each bioplastic and biodegradability.

3. Results and Discussion

3.1 Fourier transform infrared (FTIR)

Fig. 1 shows the intensity of the bond existing in the samples. The intermolecular OH vibration bond can be clearly seen in wavenumber 3200-3300cm^{-1}with a broad shape. The intensity of the O-H bond is 51-55%. Also, there is the existing C-O stretching vibration at the wavenumber 990-1060cm^{-1} with intensity of 20-29%. Nonconjugated C=C is formed at 1620-1680cm^{-1} with the intensity of 81-86%. The Formates C-O-C vibration in Ester formed at 1150cm^{-1} with an intensity of 65-69%. According to Mutmainna, I. et al. [14], starch is a type of matrix that cannot be completely bonded with the molecules of chitosan. This causes an impact on the widening of C-O peak and the intensity of C-H bond increases. The widening and shifting of the C-O peak to a lower wavenumber shows that a higher oxygen bond exists. This will result in increasing the mechanical properties of bioplastics. Samples 5 and 9 show a dominant starch, which causes the widening peak of C-O bonding and decreasing intensity of the C-H bond. For other samples, all the chitosan atoms are bonded with starch completely as a matrix. From the result and analysis, all the samples have only physical interaction.

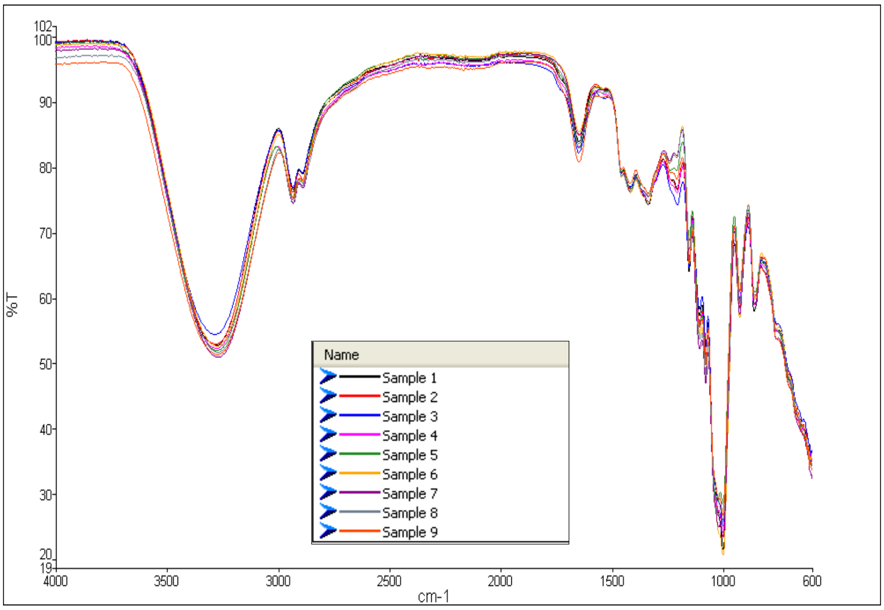
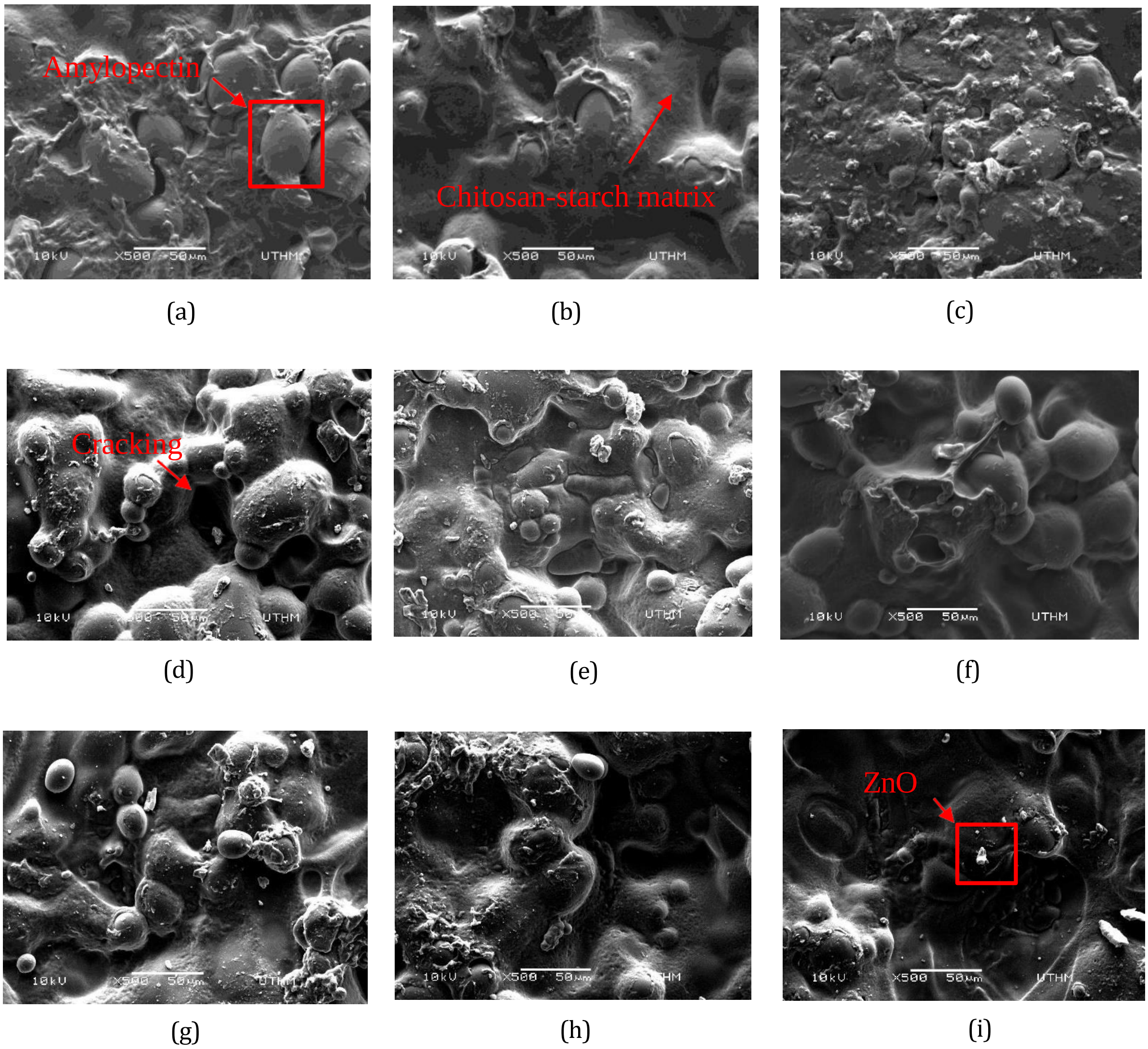


Fig. 1: *The FTIR analysis for samples 1 to 9.*

3.2 Surface morphology

The surface morphology is investigated by determining the cracks, surface smoothness and surface structure. The surface morphology of each sample is then compared to each other. By referring to Fig. 2(a), (b) and (c) obtained from Scanning Electron Microscopy (SEM), the magnification of x500 shows the obvious comparison between each sample for the surface morphology. Samples 1, 2, and 3 with the same concentration of ZnO but differ in the concentration of chitosan show that the surface of sample 3 is smoother than samples 1 and 2. The increase of Chitosan concentration makes the surface smoother due to the complete bonding of chitosan with the amylopectin. The mechanical properties are better for sample 3. For Sample 4, 5 and 6 as shown in Fig. 2(d), (e) and (f) with the ZnO of 2% (w/w) but differ in chitosan concentration show that Sample 6 surface is smoother, the cracking number is reduced but the surface is more crystalline and harder to decompose. The mechanical properties are further improved. Samples 7, 8, and 9 shown in Fig. 2 (g), (h), and (i) with the ZnO of 3%(w/w) but differ in chitosan concentration as shown in Table 1, show that the sample 9 surface is the smoothest among all samples. With the highest amount of Zinc oxide and chitosan contain, the surface is with the least amorphous. The mechanical properties are the best among all the samples, but on the other hand, these samples will take the longest time to decompose. As a result, the mechanical properties are directly proportional to the ZnO content in the bioplastics, and the higher the chitosan content, the better the degree of decomposition of the bioplastic. This statement is further supported by a previous study regarding the concentration of ZnO toward starch-chitosan based bioplastics [15].



(a) (b) (c)

(d) (e) (f)

(g) (h) (i)

Fig. 2 *SEM with 500x magnification, 50µm, 10kV. (a) Sample 1. (b) Sample 2. (c) Sample 3. (d) Sample 4. (e) Sample 5. (f) Sample 6. (g) Sample 7. (h) Sample 8. (i) Sample 9.*

3.3 X-Ray Diffraction (XRD)

The results of X-ray Diffraction show that the characterization of variations in sample 1-9 with the main ingredients of starch, ZnO and chitosan. The crystallinity value (Cr) was calculated by using the Herman method [16] shown as in Equation (1). The highest peak was determined as the intensity of crystallinity (Icrystallinity) and the valley before the crystal intensity is determined as the amorphous intensity (Iamorphous). The crystallinity value can be determined by using Equation (1). In all samples from 1 to 9, the vivid peak of the sample was at 22.15°. This result shows that the increase in ZnO concentration and Chitosan concentration does not affect the crystallization process of the bioplastics since there is no shifting of peak [3]. The valley of all samples is located at 11° to 13° [17]. The count of the vivid peak and valley for all samples is then determined and inserted into Equation (1) for calculation as shown in Table 2. Based on the crystallinity result from the table, the samples with the lower crystallinity will have better amorphous. Better amorphous samples can be considered better for decomposing compared to other samples with higher crystallinity value [17,18]. Sample 3 has the lowest crystallinity value, which represents that this sample is the easiest to decompose. The following samples are sample 1 and sample 4. Samples 7, 8, and 9 with 3%(w/w) of ZnO gave the highest crystallinity value. This suggests that a higher ZnO content increases the crystallinity of the bioplastic, which makes them more rigid and harder to decompose.

Table 2 *Crystallinity of bioplastics*

Sample	Crystal intensity (Cs)	Amorphous intensity (Cs)	Crystallinity (%)
1	870	285	75
2	996	310	76
3	863	304	74
4	933	314	75
5	988	298	77
6	912	289	76
7	927	241	79
8	738	220	77
9	979	262	79

4. Conclusion

The Bioplastics samples 1-9, which were prepared by using a combination of potato starch as matrix, glycerol, vinegar, Zinc oxide, and Chitosan, were synthesized successfully. The SEM image shows that as the Zinc oxide concentration increased; the bioplastic surface morphology became smoother. Also, the increase of chitosan concentration makes the amylopectin combine completely and form a complete matrix structure. Thus, the higher the concentration of Zinc oxide and chitosan, the better the mechanical properties such as tensile strength but lower in biodegradability. The XRD result shows that sample 3 with 74% crystallinity, which is the most amorphous, followed by samples 1 and 4 with 75% crystallinity. Samples 7 and 9 have the highest crystallinity value. Biodegradability was established for all samples that can be trimmed by microorganisms due to the presence of the functional ester group. The bioplastics samples obtained have an amorphous structure.

Acknowledgement

This research was supported by Universiti Tun Hussien Onn Malaysia (UTHM) through Tier 1 Vot Q889.

Conflict of Interest

Authors declare that there is no conflict of interest regarding the publication of the paper.

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