Synthesis of biodegradable plastics using corn starch and corn husk as the fillers as well as chitosan and sorbitol

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Abstract. Bio plastics are polymers prepared from renewable materials. In this study, maize-derived cornstarch and milled corn husk were used as the base material and filler, respectively. Corn husk powder with two-grain sizes of 150 mesh and 200 mesh, respectively, were used. Chitosan was used at concentrations of 0.02 %, 0.04 %, 0.06 %, 0.08 %, and 0.1 % by weight at a constant ratio of 1:1 to cornhusk powder and maize for improving the mechanical properties of bio plastics. The mixture was diluted using a solution containing 2.5 mL of acetic acid (25 %), 1.75 mL of sorbitol, and 70 mL distilled water. Optimum mechanical properties were observed using a cornhusk grain size of 150 meshes with 0.04% of chitosan by weight. This sample exhibited a tensile strength of 11.7164 MPa, elongation of 10.05 %, a Young’s modulus of 1.1668 MPa, and tear strength of 763.86 mN. A biodegradability of 70–100 % was achieved in 21 days with the evidence of fungal growth after 14 days. In addition, the sample was able to withstand a temperature of 140 °C for 1 h.

Keywords: bioplastic, corn husk, chitosan, sorbitol, corn starch

1. Introduction

There has been an increase in the requirements for plastics because of its cost-effectiveness, lighter weight compared to other materials such as metals and ceramics, waterproof nature, and strength. In addition to these advantages, plastics exhibit disadvantages of difficulty in rapid decomposition and non-renewability, possibly leading to environmental pollution.

A general solution to solve this issue involves the use of biodegradable polymers as the main ingredient in preparing plastics, affording bio plastics. One of the biodegradable polymers that can be used is cellulose, which is a common natural polymer. In this study, the production of such bio plastics using cellulose extracted from corn husk, which is an abundant byproduct obtained from corn processing, is investigated. Sorbitol, starch, and chitosan are used as the plasticizer, matrix, and preservative, respectively.

The use of cellulose in composite materials can lead to the increase in tensile strength owing to additional forces between molecules [1]. Sorbitol is used as a plasticizer for improving the mechanical properties of the resulting bio plastics. Generally, tensile strength increases with the molecular weight [2]. The molecular weight of sorbitol is 182.17 g/mol; this molecular weight is greater than those of other commonly used plasticizers [3].
Chitosan is mainly used as a preservative, as well as for improving the number of hydrogen bonds in the polymer chain. Hydrogen bonds contribute to the overall strength of the bioplastic [4]. Chitosan is mainly obtained from the chitinous shell of the marine crustacean exoskeleton [5].

2. Experimental method
Corn husk powder (150 mesh and 200 mesh grain sizes), powdered cornstarch, chitosan, acetic acid (25%), and distilled water were used.

First, corn husk with a grain size of 150 mesh and cornstarch are mixed in a 1:1 molar ratio. Second, different concentrations of chitosan, 0.02 %, 0.04 %, 0.06 %, 0.08 %, and 0.10 %, by weight of the mixture are added. Next, 2.5 mL of acetic acid and 1.75 mL of sorbitol are added. Finally, distilled water is added until the final volume reaches 70 mL. The procedure is then repeated for the corn husk with a grain size of 200 mesh.

The final solution is stirred and heated until it becomes homogenous and thick, followed by printing it on a non-stick polytetrafluorethylene paper with a glass plate as the supporting base. The assembly is then baked for 90 min at 80 °C.

3. Results and discussion
After the bio plastic is prepared and cooled to room temperature, several tests are carried out, including the mechanical strength test, tear test, heat resistance test, FTIR analysis, microscopy analysis, soil biodegradability (accelerated degradation test), and normal condition endurance test.

3.1. Sample appearances
Surface analysis indicated that the surface of bio plastics prepared with a grain size of 200 mesh is typically smoother compared with that prepared using a grain size of 150 mesh. The bio plastic color only varies with grain size but is not different because of the same material and composition (figure 1). Among the prepared bio plastics, bio plastics containing 0.1 wt.% of chitosan are more rigid than the others because of the presence of a high number of resulting linear chains [6].

Chitosan comprises a linear chain structure of polymers, which tends to form a crystalline phase because of the regular arrangement of polymer molecules [6]. Crystalline phases can provide strength, rigidity, and hardness [6].

3.2. Tensile strength analysis
The tensile strength graph revealed that the bio plastics prepared with a grain size of 200 mesh on average exhibit a higher tensile strength (figure 2a). This result is in agreement with that reported previously where small particles exhibit high surface energy [7]. Strength increases by increasing the surface area of coated particles through stress-transfer mechanisms [7].

For all samples, the maximum tensile strength is achieved using 0.04% of chitosan by weight, which is related to the fact that chitosan increases the number of hydrogen bonds between amylase and amyllopectin molecules in the polymer chain [8]. However, at greater than 0.04% of chitosan by weight, tensile strength generally decreases, which is related to the fact that the concentration of the dissolved chitosan affects the number of intramolecular interactions of hydrogen in chitosan [6].

Results revealed that the maximum tensile strength (figure 2a) is achieved using 0.4% of chitosan by weight, and the maximum elongation (figure 2b) is achieved using 0.6% of chitosan by weight.
3.3. Tear strength analysis

On average, samples prepared with a grain size of 150 mesh exhibit marginally higher tear strength (figure 3) as cellulose of a large grain size can contribute to the overall tear strength and afford rougher torn edges compared to samples prepared with a grain size of 200 [10,11] This result can be verified in figure 4.

3.4. Heat resistance test

All samples are subjected to heating at 100–160 °C for 60 min at intervals of 20°C. On average, all samples exhibit a weight loss between 13 % and 15 %, corresponding to the evaporation of water from
Figure 4. Samples after the tear test on the bio plastics grain size of a corn husk powder of (a) 150 mesh (b) 200 mesh

Figure 5. Physical changes of bio plastics with temperature of (a) T = 120°C, (b) T = 140 °C and (c) T=160°C

Figure 6. FTIR spectra of cornstarch (blue) and bio plastics with corn husk powder of 150 mesh (green) and 200 mesh (magenta)

The physical changes in bioplastics with respect to temperature are shown in figure 5.

3.5. Fourier transform infrared (FTIR) spectroscopy analysis

FTIR spectra of corn husk powder, maize, and bio plastic powder are shown in figure 6. Functional groups are observed for the grains of corn husk powder, cornstarch, and chitosan. In the FTIR spectrum of corn husk powder, a hydrogen-bonded O–H group is observed. The presence of CH and -CH groups revealed that corn husk comprises an insoluble nonpolar compound in water [12]; C=O, C=C, and =CH, which is a nonpolar compound insoluble in water [13], but it can dissolve in water if there is a halogen compound. FTIR results revealed that maize comprises a hydroxyl group, CH, C=C, and =CH, which are nonpolar compounds that are insoluble in water, and maize comprised a CO (ester), which permits the degradation of starch [14].

In the FTIR spectrum of chitosan, different functional groups of corn husk powder and maize are observed, including N–O (nitro) and C–N (amine). In the FTIR spectrum of sorbitol, hydrogen-bonded O–H, C–H, C=C, C=O, and C=O.
Figure 7. Microscopic examination under 50x magnification for bio plastic sample with (a) 150 mesh and (b) 200 mesh corn husk grain size

Figure 8. Degradation of bio plastics sample in (a) 1 week, (b) 2 week and (c) 3 weeks

FTIR results obtained for the bio plastic prepared using corn husk powder with grain sizes of 150 mesh and 200 mesh revealed that new functional groups are not formed; hence, the preparation of bio plastics simply involves physical blending [15]. The absorption at 2355.8 cm\(^{-1}\) is observed for bio plastics prepared using corn husk powder with a grain size of 150 mesh. It is shifted to 2359.15 cm\(^{-1}\) with a grain size of 200 mesh, which corresponds to the C=O (carbonyl) [16]. This observation is probably related to the addition of sorbitol (figure 6).

3.6. Microscopy analysis
Upon closer inspection under a light microscope, white streaks of fibers are observed for all samples (figure 7a and figure 7b), possibly because of the uneven distribution of cellulose particles in the starch matrix. A large cellulose particle size leads to the low density of cellulose particles in the starch matrix. On the other hand, a smaller or fine particle size leads to a more dense morphological structure in the starch matrix. This result implied that small particles could distribute into the starch matrix and improve the overall mechanical properties of the bio plastics.

3.7. Soil degradation test
This test aims to investigate the time required for the complete degradation of bioplastics under accelerated conditions, i.e., when samples are subjected to soil burial treatment, which is common under landfill conditions.

On average, all samples exhibit complete or near complete degradation in 30 days after continuous soil immersion. Within the first week, color and shape changes are evident, with a majority of the samples exhibiting curling. After 2 weeks, more advanced degradation is evident, with some samples already exhibiting similar soil features. After 3 weeks, a majority of the samples are completely degraded, i.e., indistinguishable from soil, with an extremely low amount of the remaining sample (figure 8).

3.8. Normal condition endurance test
The normal condition endurance test aims to determine the time required to achieve the partial or complete degradation of bioplastics under normal conditions. This test is crucial because in a commercial setting, plastics are often not used for a considerable amount of time; therefore, endurance under this condition is key to ensure that bioplastics will not degrade before time, i.e., degrade before their intended use.
Within the first week, most samples exhibit weight loss caused by water evaporation. After 2 weeks, patches of fungal growth are clearly observed on a majority of the samples (figure 9). After 3 weeks, fungal growth is subjectively more advanced, leading to mass loss on a majority of the samples.

4. Conclusions
In summary, based on the tests carried out on the samples, bioplastics prepared with a grain size of 200 mesh generally exhibit a smoother surface texture, which is possibly advantageous to the appearance of the final products. Optimum mechanical properties are achieved for the sample prepared with a grain size of 150 mesh and 0.04 % of chitosan by weight, affording a tensile strength of 11.7164 MPa, an elongation of 10.05 %, a Young’s modulus of 1.1668 MPa, and a tear strength of 763.86 mN. On average, samples prepared with a grain size of 200 mesh exhibit a higher average tensile strength of 8.4455 MPa compared to 8.2944 MPa for the samples prepared with a grain size of 150 mesh. However, for the tear strength test, samples with a grain size of 150 mesh exhibit a tear strength of 760.64 mN, which is greater than that observed for the sample with a grain size of 200 mesh (714.94 mN). FTIR analysis revealed that new functional groups are not introduced in the as-prepared bioplastics compared to the benchmark FTIR spectra of corn husk, cornstarch, and chitosan. Overall, the aims of this study have been met with these results, and this method is viable for the large-scale production of bioplastics from corn husk. A biodegradability of 70–100 % is achieved in 21 days with the evidence of fungal growth after 14 days. In addition, the sample can withstand a temperature of 140 °C for 1 h.

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