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Physical properties and soil degradation of PLA/PBAT blends film reinforced with bamboo cellulose

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Abstract. Cellulose is known as the most abundant organic molecule that is renewable and suitable to replace synthetic polymers in the production of plastics with a formula of $(C_6H_{10}O_5)_n$. Cellulose from plants is the most excellent material for reinforcing fillers. The percentage of cellulose in bamboo is range 45-55%. In this research cellulose from bamboo were incorporated with PLA and PBAT to produce cellulose film. The cellulose film produced with different amount of cellulose which are 0%, 3%, 6% and 9%. The findings showed that PLA/cellulose with 9% content of cellulose showed a higher mass loss with 12.39%, followed by the PLA/PBAT/cellulose 9%, which was 9.69%. Meanwhile, the cellulose film with 0% cellulose content for both types of plastic showed the lower biodegradability of 0.57% (PLA/cellulose bioplastic) and 0.44% (PLA/PBAT/cellulose bioplastic). It shows that biodegradability in the natural environment is the benefits of the film with a high content of cellulose. This analysis revealed the degradation of cellulose film in the soil. Usually, conventional plastic cannot easily be biodegraded by the organisms. Based on this study, it is showed that the increase of the cellulose content also encourages the film to degrade quickly. It is thus revealed that the study on bamboo's cellulose, provides the scientific information for the application and implementation of bamboo cellulose as an effective biodegradable plastics.

1. Introduction

More than decades ago, the use of plastic for packaging in everyday activities had drawn constant attention in the research community. Yet, the existing traditional methods to produce plastics are not suitable for the environment due to the increment of plastic waste [1]. Hence, to reduce the problem and these issues to the environment, the development of green product gaining recognition around the world over several years[2].

Generally, cellulose is known as the most abundant organic molecule that are renewable and suitable to replace synthetic polymers in the production of plastics. Cellulose is indeed a polysaccharide produced from glucose units and is classified as main constituent of plant cell wall along with other several substances [3]. Plus, cellulose molecules are linear and have high tendency to develop intra and intermolecular hydrogen bonds [4].

In Malaysia, *Bambusa vulgaris* (bamboo) as a lignocellulosic materials comprises of a high content of cellulose, hemicelluloses and lignin. This composition is chemically and physically bound to each other, thus produced a complex structure. Meanwhile, the minor constituents in the bamboo including



the resin, waxes, inorganic salts and tannins. The remaining 2 to 10 percent are composed of compound called extractives [5][6]. High cellulose density in the bamboo is definitely a possible alternative source for dissolving pulp processing. Bamboo cellulose have thus been used in the bioplastics production. Moreover, cellulose from bamboo becoming more common due to the excellent characteristics of bamboo in terms of degradable ability [7]. In addition, bamboo naturally is very biodegradable and versatile [7]. Hence, it can be served as a cellulose source. Consumers and industries have been interested in bamboo cellulose for the production of bioplastics as they are recognised as cheaper materials, readily available and frequently used in Asia [8].

Cellulose from plants has been proven to be the most excellent material for reinforcing fillers [8]. Cellulose bioplastics technology also has a lesser impact on the environment as it is generated from the natural-based materials that do not contain dangerous toxic chemicals [9]. Because of its potential, cellulose film from bamboo can be one of the innovative ways to replace conventional polymers in most of the plastic applications as a reinforcement [10].

A film made from the cellulose of bamboo can help to minimise environmental problem due to the excess use of conventional plastics [11]. This study aims to use bamboo as a material for the development of green plastics. Therefore, the production of cellulose bioplastics has been recommended to obtain the influence of bamboo cellulose on the physical and mechanical properties of bioplastics. Polylactic Acid (PLA) and Polybutylene Adipate Terephthalate (PBAT) also added into this research to improve the properties of the film. PLA is a well-known biodegradable polymer that has ability to function as an alternative materials while PBAT was among the plasticiser which could have been used to produce bioplastic. PBAT is biodegradable when it is thrown away due to the presence of butylene adipate groups[12]. This combination supposedly give the positive impact to the environment and the community.

2. Material and methods

2.1 Sample preparation

Bamboo (*Bambusa vulgaris*) has been collected from Agropark in Universiti Malaysia Kelantan. Bamboo was chipped into a wood chip using a wood grinder machine. Next, bamboo was undergoing a drying process in a hot oven at 50 °C for 24 hours before becoming powder. In this study, Polylactic Acid (PLA) and Polybutylene Adipate Terephthalate (PBAT) was used as a different type of plasticizer to improve the properties of film.

2.2 Preparation of cellulose from bamboo

Cellulose was prepared by mixing 100g of an oven-dry bamboo chip, sodium hydroxide (NaOH) and sodium sulfide (Na₂S) [13][14][15]. The proportion of NaOH and Na₂S required in the pulping process was calculated based on the weight of bamboo chip with sulphide and alkaline. First, NaOH and Na₂S were boiled into white liquor at a ratio of 1:8 (bamboo chip: liquid). Next, the bamboo chip was immersed in the liquor on the hot heating mantle, generally from 135 °C to 175 °C for several hours and formed bamboo pulp. The mixture was cooled, and 60 mL of distilled water was added to eliminate the traces of nitric acid. Samples then are filtered and rinsed with distilled water and ethanol. Cellulose produced at that time has been dried in the oven at 60 °C for 24 hr. Figure 1 showed the preparation of cellulose from the bamboo.

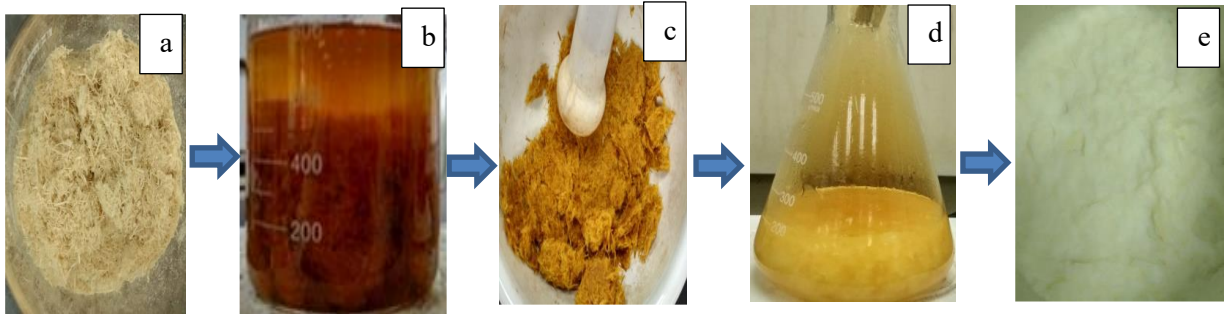


Figure 1. Preparation of cellulose from bamboo; a) Bamboo powder; b) Kraft pulping; c) Bamboo pulp; d) Alkaline treatment; e) Cellulose

2.3 Preparation of cellulose film made of bamboo

Poly(lactic acid) (PLA) and poly(butylene adipate terephthalate) (PBAT) was diluted together with a ratio of 70:30 respectively in the 100 ml chloroform for the preparation of cellulose film. Then, 15ml of the mixture was added into a beaker. The role of PBAT was to add flexibility on the bioplastics as a plasticiser. Different concentration of cellulose solution (0, 3, 6 and 9%) was added to the beaker. Then, the mixture was poured into a flat mould and dried under the room temperature for several hours then peel off from the mould. The cellulose film produced were analysed using FTIR, water absorption and biodegradation test.

2.4 Fourier Transform Infrared Spectroscopy (FTIR)

Functional group, as well as its chemical composition of cellulose film derived from bamboo cellulose, were analysed using Fourier Transform Infrared Spectroscopy (FTIR). The cellulose film samples should be made translucent using laser and infrared energy by attaching a standard scanning range of 4000 cm^{-1} to 400 cm^{-1} into the optical path [16].

2.5 Water absorption

Water absorption analysis was carried out according to ASTM D 570. The specimens with a dimension of $10\text{ mm} \times 10\text{ mm}$ were used for the water absorption test. Water absorption testing was conducted for 24 hr immersion of cellulose film in water [17]. The sample tests were reconditioned to reveal the presence of water-soluble matter in the bioplastics after 24 hr. Results of the water absorption was calculated using the following Eq. (1).

$$\text{Water absorption percentage (\%)} = \{(W_f - W_i) / W_i\} \times 100\% \quad (1)$$

Where W_i is the weight of the dry sample, and W_f is the weight of the sample immersed in distilled water for 24 hr.

2.6 Biodegradable test.

Samples was cut into the dimension of $40\text{ mm} \times 40\text{ mm}$. 500g of the soil containing abundant nitrogenous bacteria was collected and safely stored in a container. Each cellulose film was buried inside the soil at 3 cm depth for 15 days under the room conditions to identify possible degradation. Weight of specimen was taken before and after the test. Samples are weighed 15 days to evaluate the percentage of mass loss. Results were obtained based on the Eq. (2).

$$\text{Mass loss (\%)} = \{(\text{Initial mass} - \text{Final mass}) / \text{initial mass}\} \times 100 \quad (2)$$

3. Results and Discussions

3.1 Functional group of a cellulose-based film made of bamboo

Figure 2 showed the FTIR spectra of the cellulose film with different percentage of cellulose which is of 3%, 6% and 9%. At the range of wavelength 3355 cm^{-1} to 3358 cm^{-1} shows the presence of OH groups, which is part of the functional group of cellulose [16]. This also was supported by the existence of typical groups of cellulose other than the hydroxyl group, such as the methylene group (CH₂) in the range of 2945 cm^{-1} to 2955 cm^{-1} absorption band which was a C-H stretch vibration.

The results also dealt with the presence of a medium peak at range of 1267 cm^{-1} to 1269 cm^{-1} that was due to C-O stretching, which described the stretching of cellulose. Lastly, the small sharp peaks around 753 cm^{-1} showed that samples with 6% and 9% of cellulose content have a higher strong peak of C-H bending compared to the other samples. This result was assigned to the linkage between the sugar units in the cellulose.

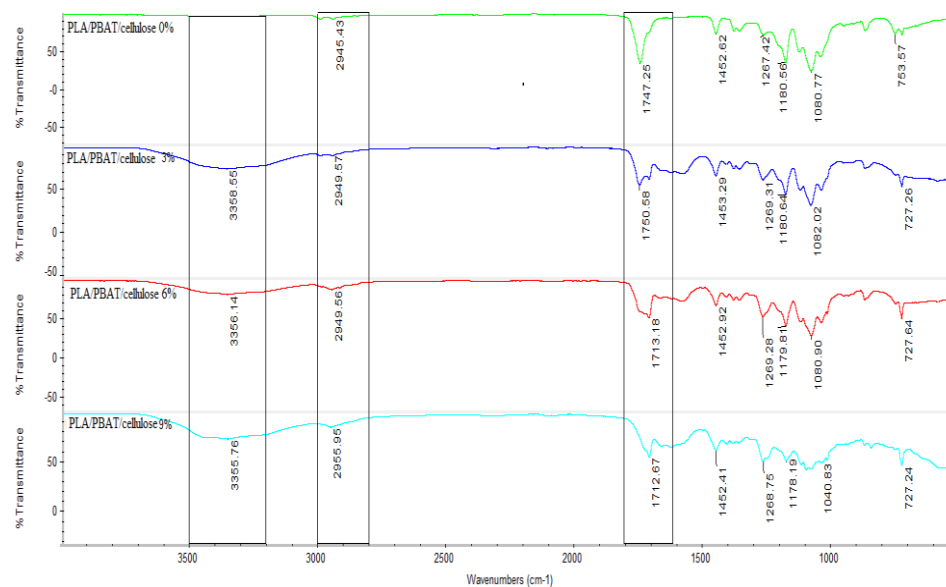


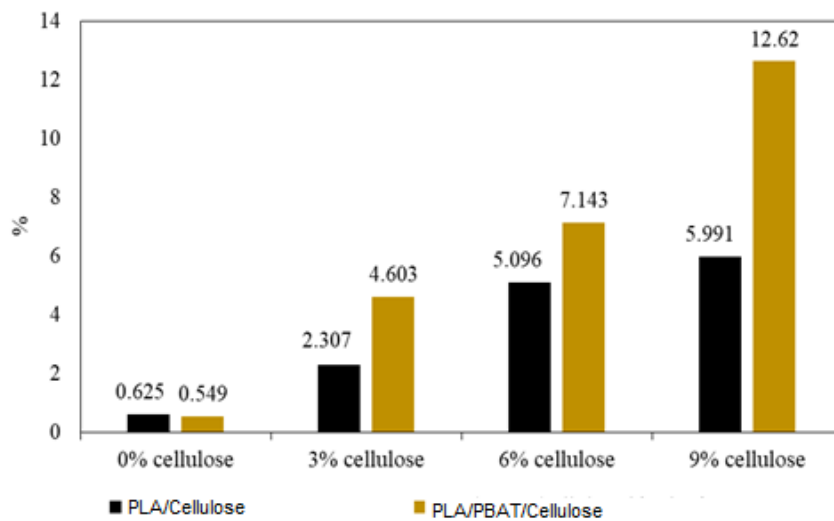
Figure 2. The functional group of cellulose film made of bamboo

3.2 Water absorption of cellulose film made of bamboo

Water absorption test is beneficial in determining the water absorptivity of the cellulose film. Figure 3 revealed the water absorption of the cellulose film by a different percentage of cellulose. From the figure, the water absorption rate of the cellulose film increased with a higher content of cellulose due to the properties and characteristics of cellulose itself. The increment of water absorption to cellulose and fibre loading suggests that cellulose extracted from this experiment are somewhat hydrophilic. The findings showed that PLA/PBAT with 9% cellulose film sheets had the highest water absorption percentage trailed by PLA/PBAT with 6% cellulose film sheet. Increasing water absorption also can be improved by adding a small amount of fibres [17].

Moreover, the cellulose film sheets PLA/cellulose 0% and PLA/PBAT/cellulose 0% had the minimum water absorption rate which was 0.625% and 0.529% respectively and followed by cellulose film sheet with 3% cellulose. This results might be due to the pure polymer content in the cellulose film. Besides, it can be concluded that the maximum content of cellulose in cellulose film sheets with 9% of cellulose showed the highest water absorption for both types of plastic. The high amount of cellulose in the cellulose film was the main factor of the high percentage of water absorption. The appearance of hydroxyl groups in cellulose fibres interrelates with the water molecules which encourage the intake of water [18]. PLA and PBAT itself is hydrophobic and not susceptible to the moisture, therefore presents low water absorption. But, it was not sufficient to enhance the repellency of water absorption [19].

Hence, it concluded water of cellulose to increase cellulose increased.



can be that the absorption film tends when the content

Figure 3. Rate of water absorption in cellulose film

3.3 Degradation of cellulose film made of bamboo

Degradation test proved the microbial activity on the cellulose film samples. Based on Figure 4, PLA/cellulose with 9% content of cellulose showed a higher mass loss with 12.39%, followed by the PLA/PBAT/cellulose 9%, which was 9.69%. Meanwhile, the cellulose film with 0% cellulose content for both types of plastic showed the lower biodegradability of 0.57% (PLA/cellulose bioplastic) and 0.44% (PLA/PBAT/cellulose bioplastic).

The result reveals that changes in mass loss of cellulose film relying on the variations of cellulose percentage. Highest biodegradability of 12.39% was recorded in 15 days for the PLA/cellulose film with 9% cellulose content. It may be due to the properties of cellulose film itself, which easily degraded due to the amorphous nature of the polymer [20][21]. Therefore, the microorganisms in the soil could invade the bioplastic molecules [21]. In reality, cellulose from a natural substance is believed to be biodegradable, and the research study reported that almost 90% of cellulose could decompose within 6 months and it can be accomplished in less than 45 days with industrial composting [21].

On top of that, combination of this formulation also claimed as compostable polymer. They have been known as thermoplastics and helped cellulose film degrade fastly in this study. As a result, it may be concluded that the combination of the thermoplastics with a higher content of cellulose (9%) can accelerate the degradation of the cellulose film. Thus, the mass loss of cellulose film increased as the cellulose content increased. Figure 5 represented the images of cellulose film after 15 days.

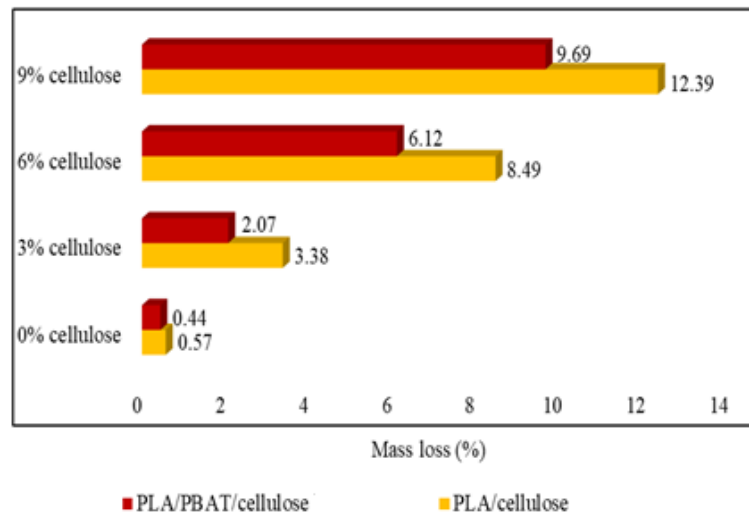


Figure 4. Mass loss of the cellulose film after 15 days

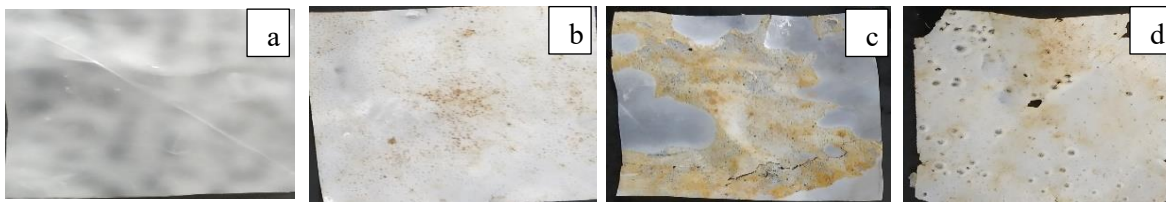


Figure 5. Images of cellulose cellulose film after 15 days. (a) PLA/PBAT/cellulose 0%, (b) PLA/PBAT/cellulose 3%, (c) PLA/PBAT/cellulose 6%, (d) PLA/PBAT/cellulose 9%

4. Conclusion

Introducing cellulose film to the biodegradable plastics helps to promote and improve soil degradation. The degradability in the natural environment is the benefits of bioplastic with a high content of cellulose. This analysis demonstrated the degradation of cellulose film in the soil. Microorganisms could degrade cellulose film in the soil. On the other hand, conventional plastic cannot easily be degraded by the organisms. Based on this study, it is showed that the increase of the cellulose content also encourages the plastics to degrade quickly. It is thus revealed that the study on bamboo's cellulose, provides the scientific information for the application and implementation of bamboo cellulose as an effective biodegradable plastics.

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