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High-Content Lignocellulosic Fibers Reinforcing Starch-Based Biodegradable Composites: Properties and Applications

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Abstract

Natural source-based composites became promising substitutes and synthetic petrochemical-based counterparts. So far, thermoplastic starch and lignocellulosic fibers are the most common materials for making such eco-friendly “green” materials. Low cost, abundance, and renewability are the factors that lead to deploying these two types of materials. In this chapter, we are conducting further analysis for previously published results of six types of high-content natural fiber-reinforced starch-based composites. All composites were prepared by compression molding under pressure from 5 to 20 MPa and temperature from 130 to 160°C. Composites exhibited highest tensile strength and modulus of elasticity at fiber weight content from 50 to 70%, and then mechanical properties deteriorated significantly at 80% fiber content due to the insufficient starch resin. For instance, the tensile strength was boosted up from 2-12 MPa for thermoplastic starch to reach 55, 45, 32, 28, 44, 365 MPa for flax, bagasse, date palm fiber (DPF), banana, bamboo, and hemp composites, when fiber content was increased from 0% to the optimum fiber content (50-70%). Kelly-Tyson (random 2d) was the optimum model to predict random fiber composite. Increasing the fiber content and choosing a fiber with high cellulose content significantly improve the moisture resistance of the composites. Fick’s law of diffusion predicted the water uptake property successfully. The thermal stability of composites was improved with increasing the fiber weight content as well. This is attributed to the high thermal stability of cellulose when compared to starch. Properties exhibited by starch-based high-content natural fiber composite are promising for many industrial and biomedical applications.

Keywords: natural fibers, composite, mechanical properties, water uptake, thermal properties, theoretical models

1. Introduction

In the past two decades, environmentally friendly biodegradable plastics have been receiving attention because of the need to diminish the worldwide pollution caused by petroleum-based synthetic plastics [1, 2]. Biodegradability by definition corresponds to the capacity of the material to be completely assimilated by indigenous micro-organisms in the ecosystem. Every year more than 300 million tons of plastics are produced for different applications [3]. In addition to nonrenewability of the synthetic plastics resource, disposing of those types of plastics in landfill will inevitably release toxins in the soil and underground water. Moreover, contaminants can be absorbed in food resources and eventually accumulated in the human body [4, 5].

Bioplastics from natural resources provide reliable and sustainable alternative to synthetic polymer. Starch is the most commonly studied materials as an eco-friendly polymer that is based on renewable plant material, fully biodegradable, and low cost [6, 7]. There are some challenges that oppose the development of starch-based plastics. Those challenges are mainly poor long-term stability, low water resistance, deterioration of mechanical properties due to moisture uptake, and the relatively fast biodegradability [8]. To overcome those challenges, natural lignocellulosic fibers have been widely utilized to improve the properties of the starch-based plastics [9]. Comparable to synthetic fibers, natural fibers are less dense in addition to being fully degradable. Furthermore, the reinforcement of lignocellulosic fibers significantly improves the mechanical properties of starch-based matrix [10]. Sisal, jute, kenaf, coir, wood, pulp, cellulose, bagasse, banana, orange, and flax fibers are lignocellulosic fibers that have been all studied and found to substantiate enhancement for the starch-based matrix's versatile properties [10–14]. The objective of this article is to review and conduct further analysis to understand the effect of high-fiber content (flax, palm, banana, bagasse, bamboo, and hemp) on mechanical and physical properties. Moreover, experimental results reported before will be compared with theoretical models, selecting the most appropriate model.

1.1. Preparation and characterization

For flax, bagasse, banana, and date palm fiber (DPF) preparation took place by preheating the composite, pressing at 5 MPa and 150°C for 30 min [13–16]. Bamboo-reinforced starch composite was prepared by pressing at 20 MPa and 130 C for 5 min [17]. Hemp-reinforced starch composite was prepared by pressing at 10 MPa and 130 C for 5 min [18]. All fibers except bamboo and hemp were subjected to alkaline (NaOH) treatment before composite preparation. In this chapter, we will deal with tensile, moisture, and thermal results, and further details concerning experimental procedures can be found in the cited articles.

2. Results and properties

2.1. Tensile properties

2.1.1. Strength

Role of mixture (ROM) and inverse role of mixture (IROM) are the most common theories to predict composite mechanical behavior. ROM assumes that there is a perfect bond between

fibers and matrix, also that the strain (and stress for IROM) in the fibers is equal to the strain in the matrix. This model deals with aligned continuous fiber composites [19, 20].

ROM equation predicts tensile strength in longitudinal direction:

$$\sigma_L = \sigma_F V_F + \sigma_M V_M \quad (1)$$

IROM equation predicts tensile strength in transversal direction:

$$\sigma_T = \frac{\sigma_F \sigma_M}{V_M \sigma_F + V_F \sigma_M} \quad (2)$$

where σ_F is the fiber tensile strength, σ_M is the matrix tensile strength, V_F is the fibers volume fraction, and V_M is the matrix volume fraction. In order to transform fiber weight fraction (W_F) to volume fraction (V_F), we will be in need to the following equation:

$$V_F = \frac{\rho_M W_F}{\rho_M W_F + \rho_F W_M} \quad (3)$$

where ρ_F is the fiber density, ρ_M is the matrix density, and W_M is the matrix weight fraction.

Kelly-Tyson formulates an approach to deal with discontinuous (random and unidirectional) fiber, overcoming the problem of unequal strain in the fibers and the matrix by assuming perfect bonding between fibers and matrix. If the fiber is shorter than the critical length (L_c), it cannot be loaded to its failure stress, and the strength of the composite is then determined by the strength of the fiber/matrix bond, in addition to fiber strength. So for fibers' length greater than L_c , the composite tensile strength can be calculated as follows [19]:

$$L_c = \frac{\sigma_F d}{2\tau} \quad (4)$$

$$\sigma_C = \varepsilon \left(1 - \frac{L_c}{2L} \right) \sigma_F V_F + \sigma_M V_M \quad (5)$$

where L_c is the critical length, d is the fibers' average diameter, τ is the shear strength of fiber/matrix bond (about 0.5 of matrix strength), ε equals 1 in case of unidirectional distribution and three-eighth in random distribution. L is the fibers' average length, σ_F is the fibers' tensile strength, σ_M is the matrix tensile strength, V_F is the fibers' volume fraction, and V_M is the matrix volume fraction. Different properties have been investigated from low to high fiber weight content (50–80%; **Figure 1**). For all types of fibers, mechanical properties improved as the fiber content increased from 0 to 50–70%. Strength of 0% (starch matrix) was around 4 MPa for the flax, bagasse, and DPF. Starch matrix recorded lower strength in the case of banana composite (2 MPa), whereas starch-based resin strength used in bamboo and hemp composites was 12 MPa. Among the first four types of composites, 50–60% fibers' composite was 8–15 times higher than 0%, depending on the fiber type. Flax composite exhibited the highest mechanical properties with tensile strength surpassing 60 MPa, banana composite was the lowest in strength (27 MPa). About 80% composite was flimsy with

Figure 1. Experimental and theoretical tensile strength of starch-based lignocellulosic fiber composites for different fibers: (a) flax, (b) bagasse, (c) DPF, (d) banana, (e) bamboo, and (f) hemp.

severely deteriorated tensile properties due to lack of matrix material; hence, the load cannot be transferred from fiber to another. For the last two types, the trials did not continue to recognize the maximum possible fiber content for maximal strength (no reduction in strength after maximum strength point). Hemp fiber composite recorded 365 MPa for 70% of the fibers, the high strength of hemp fiber (above 700 MPa), and availability of hemp in long bundles are reasons for this high strength. Kelly-Tyson's (random 2d) was the closest model to experimental results for flax, bagasse, and DPF composites. In the case of banana and bamboo fibers, the experimental results were between Kelly-Tyson (random 2d) and IROM, with more inclined trend to IROM, and this can be due to higher levels of randomness (3d), especially for shorter fibers (banana). Hemp fibers were continuous fibers; hence, the results were close to ROM model.

2.1.2. Elasticity

Similar to ROM and IROM strength models, the two models predict longitudinal and transversal elasticity values for unidirectional continuous fiber composites in which the actual composite elasticity value must lie between. ROM equation predicts elastic modulus in longitudinal direction (E_L):

$$E_L = E_F V_F + E_M V_M \quad (6)$$

And IROM equation predicts elastic modulus in transversal direction (E_T):

$$E_T = \frac{E_F E_M}{V_M E_F + V_F E_M} \quad (7)$$

where E_F is the fibers modulus of elasticity, E_M is the matrix modulus of elasticity, V_F is the fibers volume fraction, and V_M is the matrix volume fraction.

Another model used to predict discontinuous fiber composite behavior was called Halpin-Tsai. The composite elastic modulus in longitudinal direction (E_L) can be obtained as follows [15, 19]:

$$E_L = E_M \frac{(1 + \xi_L \eta_L V_F)}{(1 - \eta_L V_F)} \quad (8)$$

where (ξ) is a factor dependant on the shape and distribution of the reinforcement. The parameter (η) is a function of the ratio of the relevant fiber and matrix moduli with respect to the reinforcement factor (ξ). In this model, they can be calculated by the following formulas:

$$\eta_L = \frac{(E_F - E_M)}{(E_F + \xi_L E_M)} \quad (9)$$

$$\xi_L = 2 \left(\frac{L}{d} \right) + 40 V_F^{10} \quad (10)$$

Also, the composite elastic modulus in transversal direction (E_T) can be obtained as follows:

$$E_T = E_M \frac{(1 + \xi_T \eta_T V_F)}{(1 - \eta_T V_F)} \quad (11)$$

where

$$\eta_T = \frac{(E_F - E_M)}{(E_F + \xi_T E_M)} \quad (12)$$

$$\xi_T = 2 + 40 V_F^{10} \quad (13)$$

The two previous moduli are for oriented fibers, the following equation is a result of an averaging process made by Tsai and Pagano [21] to predict the modulus of an isotropic composite (E_c) based on random fibers reinforcement.

$$E_C = \frac{3}{8} E_L + \frac{5}{8} E_T \quad (14)$$

Figure 2 shows the modulus of elasticity for the first four composite types discussed in the strength section. Young's modulus followed the same pattern as tensile strength. Modulus increased with fiber content till an optimum point (50–60%), then property deterioration took place at higher content (80%) due to the scarcity of resin and inability to transfer load between fibers. Flax composite exhibited the highest modulus (4.7 GPa) in comparison with other types. Superior mechanical properties for flax were realized elsewhere and attributed to high

Figure 2. Experimental and theoretical tensile modulus of starch-based lignocellulosic fiber composites for different fibers: (a) flax, (b) bagasse, (c) DPF, and (d) banana.

cellulose content and low fiber diameter. Halpin-Tsai was the best model to predict the composite behavior, and this is due to the randomness of the fibers used to prepare these composites.

2.2. Moisture absorption behavior

Owing to its natural abundance and low cost, starch-based biodegradable “green” polymers have attracted great attention. Unfortunately, the use of these plastics in a wide range of applications has been restricted by its low water resistance. Therefore, in order to overcome this disadvantage while preserving the biodegradability and the green property of this polymer, natural lignocellulosic fibers are being used as a biodegradable and eco-friendly reinforcement [22–25]. The incorporation of natural lignocellulosic fibers, which are mainly made up of hydrophilic cellulose, into starch-based matrix is responsible for the reduction of moisture absorption of the resultant composite. This reduction in the moisture absorption of the two hydrophilic materials is attributed to the good interfacial adhesion between starch and cellulose which leads to decreasing the free volume of the starch molecular chains and thus reduce the water absorption; the less hygroscopicity of cellulose when compared with starch; formation of fibrous network around starch thus hinder the moisture penetration; and the high crystallinity of cellulose when compared with starch [26–29].

2.2.1. Mechanism of moisture absorption in polymer/natural fiber composites

Polymer/natural fiber composites tend to absorb moisture in humid atmosphere. Therefore, this moisture absorption affects the fiber matrix interface leading to low stress transfer between matrix and fiber. The transport of water in a polymer/fiber composite can be attributed to the imperfections of the matrix (voids, pores, and cracks). The water absorbed in a matrix can be divided into two types, free water and bound water. Free water is the water molecules moving freely through the voids, and bound water is the water molecules bonded to the polar groups of a matrix [30]. **Figure 3** shows the two types of absorbed water in polymeric matrix.

When water molecules penetrate polymer/fiber composite, it attaches to the hydrophilic group of the fiber creating intramolecular hydrogen bonds. Thus, this interaction reduces the interfacial adhesion between fiber and matrix which leads to deterioration in the mechanical properties of the composite [30, 31]. **Figure 4** explains how moisture absorption by fiber affects the composite.

Polymeric matrices, especially, starch-based reinforced with natural lignocellulosic fibers are sensitive to moisture. Moisture absorption will deteriorate their functionality. Therefore, aspects should be considered when manufacturing such a composite that would be used in a humid atmosphere. The most important aspect is the proper selection of the fiber type according to its moisture resistance.

2.2.2. Effect of fiber type and content on moisture absorption

According to literature, the moisture absorption of starch/natural fiber composites is highly affected by the fiber type and its content. Each type of fibers has its own cellulose content; thus fibers with high cellulose content highly diminish the moisture absorption of the resultant composite. Mehanny *et al.* [13] studied the effect of reinforcing thermoplastic starch (TPS) matrix with different contents of NaOH-treated bagasse fiber on the moisture absorption of the resultant composite. **Figure 5** shows the moisture absorption of composites reinforced with 0, 20, 40, 60, and 80% bagasse fiber. Their results showed that the moisture absorption after reaching equilibrium of the starch-based matrix with 0% fiber reached more than 53%,

Figure 3. Free water and bound water in polymer matrix. Adopted from Ref. [30].

Figure 4. Effect of water on fiber-matrix interface. Adopted from Ref. [30].

Figure 5. Moisture absorption of TPS/bagasse fiber composites with different fiber wt%. Adopted from Ref. [13].

whereas with the lowest fiber content 20% it reached 48%. By increasing the fiber content up to 80%, the moisture absorption dropped to 36%.

According to Elsayed *et al.* [14, 15], increasing the NaOH-treated flax fiber content from 0 up to 60% resulted in reducing the moisture absorption of the TPS/flax composite from 48 to 38% as shown in **Figure 6**. The authors proposed investigating, as well, the effect of changing

Figure 6. Moisture absorption of (a) TPS/flax fiber and (b) TPS/DPF composites with different fiber wt%. Adopted from Refs. [14, 15].

the reinforcement fiber on the moisture absorption of the resultant composite. NaOH-treated date palm fiber (DPF) with different contents was used. They demonstrated that changing the fiber type caused a slight effect on the moisture absorption property of the resultant composite. The authors attributed this slight difference to the convergence of the cellulosic content of both fibers.

Darwish *et al.* [16] investigated the effect of increasing the content of the NaOH-treated banana fiber (BF) reinforcement on the moisture absorption property of the starch/BFs composites as shown in **Figure 7**. The starch matrix was reinforced with 40, 50, and 60% BFs. The moisture absorption dropped from 70 to 41% with increasing the BF content from 0 to 60%.

Figure 8 shows a comparison between the moisture absorption of the TPS composites reinforced with 60 wt% fibers at the equilibrium plateau from the previously stated studies [13–16]. From **Figure 8**, it can be concluded that the moisture absorption is highly dependent on the fiber type, hence the cellulose content of the fibers. Therefore, flax with the highest cellulose content diminished the moisture absorption of the composite to 35%.

2.2.3. Fick's law and moisture absorption of TPS/natural fiber composites

Fick's second law of diffusion has been widely used to model and characterize the absorption of moisture in many materials. Fick's second law depicts the process of one dimension moisture absorption with respect to exposure time as shown in Eq. (1) [32–35]:

$$\frac{dC}{dt} = D \frac{d^2C}{dX^2} \quad (15)$$

where C is the water concentration, t is the time of diffusion, and D is the diffusion coefficient normal to the surface in the x -direction.

Figure 7. Moisture absorption of TPS/banana fiber composites with different fiber wt%. Adopted from Ref. [16].

Figure 8. Moisture absorption of TPS composites reinforced with 60% fibers at the equilibrium plateau.

Diffusion behavior, Fickian or non-Fickian, can be recognized theoretically by the shape of the absorption curve represented by

$$\frac{M_t}{M_m} = K \cdot t^n \quad (16)$$

where M_t and M_m are moisture absorption at time t and at equilibrium state, respectively. K and n are the diffusion kinetic parameters. M_t can be calculated from

$$M_t = \frac{W_t - W_0}{W_0} 100(\%) \quad (17)$$

where W_0 is the weight of dry sample and W_t is the weight of wet sample at time t [32–35].

The diffusion parameter n indicates whether the diffusion is Fickian or non-Fickian. When $n = 0.5$, the diffusion is Fickian. When $n \geq 1$, the diffusion is non-Fickian. Moisture absorption in lignocellulosic fiber-reinforced polymer composites always follows Fickian behavior [32–36].

One-dimensional approach of Fick's law shows that the moisture absorption is directly proportional to the square root of time, then slows down until an equilibrium plateau is reached. For values of $M_t/M_m < 0.6$, the initial part of the curve can be deduced by [36]:

$$\frac{M_t}{M_m} = \frac{4}{h} \sqrt{\frac{D \cdot t}{\pi}} \quad (18)$$

where h is the thickness of the sample.

For the second half of the absorption curve, where $M_t/M_m > 0.6$, Springer [37] proposed the following approximation:

$$\frac{M_t}{M_m} = 1 - \exp \left[-7.3 \left(\frac{D \cdot t}{h^2} \right)^{0.75} \right] \quad (19)$$

Figure 9 shows a comparison between the Fick's law predicted and the experimental moisture absorption test results of TPS composites reinforced with 60 wt% of different fibers (banana, bagasse, DPF, and flax). The four curves show that the experimental data are in a good agreement with the predicted values. Thus, TPS/lignocellulosic fiber composites follow a Fickian behavior.

Figure 9. Experimental and predicted moisture absorption of TPS composites reinforced with 60% (a) BFs, (b) bagasse fibers, (c) DPFs, and (d) flax fibers.

2.2.4. Effect of moisture absorption on mechanical properties of composites

Mechanical properties deterioration is one of the drawbacks of moisture absorption on polymer/natural fiber composites. This deterioration is attributed to the swelling of the cellulose fibers. Due to this swelling, development of shear stress at the fiber/matrix interface occurs. Therefore, this leads to debonding of the fibers, delamination, and loss of structural integrity [30, 38]. Fiber surface chemical treatments are proven to promote the moisture resistance by reducing fiber hydrophilicity. Moreover, these treatments improve the interfacial adhesion between matrix and fiber thus, tightens the water penetration pathways and consequently, lowers the moisture absorption [30, 39–41].

In Ref. [25], the authors studied the effect of moisture absorption on the mechanical properties of starch-based composites reinforced with different contents of NaOH-treated flax and DP fibers. Tensile strength test results showed that the strength of both flax and DPFs composites decreased to the half of its original value as shown in **Figure 10**.

In sum, from the previously illustrated studies, moisture absorption of TPS/lignocellulosic fiber composites is affected by the type and the content of the reinforcement fiber. Moreover, different fiber surface treatment techniques highly modify its water resistance. Therefore, proper selection of fiber type, content, and surface treatment technique is of crucial importance while manufacturing a composite exposed to humid atmosphere in order to preserve its strength and structural integrity.

Figure 10. Effect of moisture absorption on the tensile strength of (a) flax composites and (b) DPFs composite. Adopted from Ref. [42].

2.3. Thermal properties

Thermogravimetric analysis (TGA) is a widely used thermal technique due to its high accuracy in determining the decomposition temperature and thermal stability of materials. TGA measures the rate and amount of weight change of a material as a function of time or temperature in a controlled atmosphere. This technique proved to be useful in studying the thermal characteristics of polymeric materials such as thermoplastics, composites, films, and fibers [43]. The thermogravimetric graph for thermoplastic starch matrix is shown in **Figure 11**. The authors in references [13–16] reported that the thermal decomposition of thermoplastic starch occurs in three main steps. The initial weight loss that occurred in the TGA curve between room temperature and 100°C represents the first step. This weight loss was attributed to the evaporation of water. It can be observed as a small peak around 100°C in the DTG curve. The second step appeared as a peak in the DTG curve around 200°C. This peak was attributed to the evaporation of glycerin. The last step appeared as a major peak around 330°C in the DTG curve. This peak was attributed to thermal decomposition of starch.

TGA and DTG curves for different lignocellulosic fibers are comparable due to their chemical composition similarity. On the other hand, chemical surface treatments affect the thermal stability of the fibers [40, 45]. Darwish *et al.* [16] studied the effect of NaOH treatment on the thermal stability of BFs. Their results showed that TGA and DTG curves were shifted to the

right after treatment (**Figure 12**). This shift indicated that the treated fiber exhibited an increased thermal stability relative to the untreated fiber.

Figure 11. (a) TGA and (b) DTG of TPS matrix. Adopted from Refs. [13, 14, 44].

Figure 12. (a) TGA and (b) DTG of untreated and treated BFs. Adopted from Ref. [44].

TGA and DTG curves were plotted by the authors in references [13–16] to analyze the thermal degradation of starch-based composites reinforced with different contents of NaOH-treated lignocellulosic fibers (bagasse, flax, DPF, and banana) (**Figure 13**). The authors reported that the decomposition of TPS/natural fiber composites has two main peak regions. From the DTG curves, peaks, Group I represents the first decomposition region which is attributed to the decomposition of starch. These peaks appeared at the temperature range of 300–340°C. The second decomposition region appeared in the DTG curves as peaks Group II in the temperature range of 340–400°C. These peaks are attributed to the decomposition of the lignocellulosic fibers.

The authors in references [11–14] reported that the small peak that is related to the evaporation of water disappeared at high fiber contents. They attributed this disappearance to the improvement of the moisture absorption resistance of the composites with higher fiber contents. Also, the authors noted that as the fiber content increases, the residual weight decreases, which imply that starch-based matrix contains more inorganic inclusions. The authors noticed that the onset degradation temperature of starch increased with increasing the fibers' content. They assigned this improvement to the possible formation of hydrogen bond linkage between starch

Figure 13. TGA and DTG curves for TPS composites reinforced with (a) flax fibers, (b) DPFs, (c) banana fibers, and (d) bagasse fibers. Adopted from Refs. [13–16, 46].

and fibers. The mentioned studies [13–16] reported that with increasing the fiber content, the onset degradation temperature of the composites increased and the weight loss decreased. Thus, the thermal stability of the composites improved with increasing the fibers content. This improvement was attributed to the higher thermal stability of the cellulosic fibers when compared with starch and the good compatibility of both polysaccharides. Moreover, proper selection of the surface treatment technique of the fibers plays a prominent role in improving the thermal stability of the resultant composite.

In short, the parameters obtained from the thermal characterization of TPS/lignocellulosic fiber composites are useful in identifying the temperature limits of treatment, processing, or operating both matrix and fibers. Moreover, these parameters are useful in comparing the thermal performance of various TPS/lignocellulosic fiber composites.

3. Conclusion

- For flax, bagasse, DPF, and banana composites, samples with 50-60 wt% fibers manifest the highest tensile strength of 8-15 times what was recorded by 0% composite (less than 4 MPa strength for 0% fiber content).
- In 80 wt% composite, insufficient matrix is unable to transfer load and fill the gaps between fibers, leading to severely deteriorated tensile properties.
- Kelly-Tyson (random) model shows the best fit for the mentioned composites, which can be attributed to the realistic assumptions that count for the randomness of the fibers during preparation.
- For hemp composite, strength can be increased to 365 MPa, when increasing fiber content to 70%. Small diameter and continuous fibers with such high strength (702 MPa) led to the significant composite strength improvement.
- ROM model can be used for tensile strength and modulus prediction for hemp composites in case of fiber continuity in the composite.
- Halpin-Tsai (random) is the most suitable model to predict starch composite behavior.
- The moisture absorption of TPS/lignocellulosic fiber composites is affected by the fiber type and content. Increasing the fiber content and choosing a fiber with high cellulose content improve the moisture resistance of the composites. Moreover, proper selection of the surface treatment technique of the fiber highly modifies its water resistance.
- The moisture absorption behavior can be modeled successfully by Fick's law of diffusion.
- The thermal stability of the TPS/lignocellulosic fiber composites is improved with increasing the fiber content. This is attributed to the high thermal stability of cellulose when compared to starch. Furthermore, proper selection of the surface treatment technique of the fiber enhances the thermal stability of the fiber itself and the resultant composite.

- To further improve moisture resistance properties, hydrophobic modified starch, or hydrophobic coating can be implemented.
- Starch-based lignocellulosic fiber can be a good candidate for regenerating broken bones (producing bone plates) due to the biocompatibility of starch plastics (especially after modifying the moisture uptake issue).
- Starch-fiber panels can be a good candidate for many paneling applications relevant to housing, cars, and decorative applications.

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