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RESEARCH ARTICLE

Preparation of Microcrystalline Cellulose from Bagasse Bleached Pulp Reinforced Polylactic Acid Composite Films

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Abstract Polylactic acid (PLA) is biodegradable and biocompatible, but some of its functional properties limit its commercial application. To improve the properties of PLA without destroying other useful characteristics, a composite film consisting of microcrystalline cellulose (MCC) and PLA was prepared. In this research, MCC was obtained from bagasse bleached pulp by hydrochloric acid hydrolysis. After being modified by a silane coupling agent (KH550), the MCC was used as reinforcement in the PLA matrix. The degradation performance of the composite film was explored using degradation experiments in which the film was buried in soil. According to the single-factor experiments, the best composite formulation was 3% KH550, 20% KH550-MCC and 15% acetyl tributyl citrate. The results showed that the interfacial compatibility of the MCC/PLA composite films was improved after MCC was modified with KH550. This research showed that the addition of MCC can enhance the degradation rate of MCC/PLA composite films.

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Keywords Polylactic acid · Microcrystalline cellulose · Composite membrane · Degradation

Introduction

With the reduction of petroleum resources and the growth of environmental awareness worldwide, many researchers have been motivated to develop bio-based materials to replace traditional petrochemical-based materials (Farah et al. 2016). Among these candidates, polylactic acid (PLA) has gained much attention and is now widely regarded as the most promising new "ecological material" of the twenty-first century (Suryanegara et al. 2009). PLA is a biodegradable and thermoplastic aliphatic polyester that is derived from renewable resources such as corn, potato starch and other organic sources (Jonoobi et al. 2010). Besides its biological characteristics, PLA also exhibits excellent mechanical strength, which makes it widely used in packaging, drug delivery products and biological scaffolds (Kamal and Khoshkava 2015).

However, slow degradation rate (Tokiwa and Calabia 2006), brittleness (Suryanegara et al. 2009) and poor thermal stability (Södergård and Stolt 2002) limit the wide application of PLA without modification. A possible solution to this problem is to combine PLA with some micro- or nanoscale materials (Armentano et al. 2013); one such material is microcrystalline cellulose (MCC). MCC is a fine, odorless, crystalline powder that is biodegradable and can be degraded by natural cellulose; because of these properties, MCC has been used in the pharmaceuticals, food and cosmetics fields (Mohamad Haafiz et al. 2013). To reduce the cost of producing MCC, researchers have focused on inexpensive raw materials, such as waste cotton

(Chuayjuljit et al. 2009), corn stover (Zhang et al. 2012), bagasse (Ilindra and Dhake 2008) and other types of biomass. MCC consists of small particles that have large specific surface area, leading to the formation of intermolecular hydrogen bonds among the hydroxyl groups on the particle surface and subsequently causing agglomeration (Adel et al. 2011). Key to reinforcing the mechanical and thermodynamic properties of PLA is improving the compatibility between PLA and MCC. Silane coupling agents have been used to solve the compatibility problem (Cui et al. 2018).

The objectives of this study were to provide a simple, efficient and innovative method to modify and improve the performance of PLA and provided experimentally determined guidance for producing PLA-based composite materials. In this research, MCC was prepared from bleached bagasse pulp using hydrochloric acid hydrolysis. The MCC was modified using a specific silane coupling agent (KH550), and modification conditions were explored using single-factor experiments. Finally, the modified MCC was incorporated into PLA using a melt mixing method during which the preparation process of composite films was investigated. Scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) and thermogravimetric analysis (TGA) were used to characterize the bleached pulp, MCC, modified MCC and the prepared composite films. The mass and surface changes of the composite films were explored through degradation experiments in which the material was buried in soil.

Materials and Methods

Material

Bagasse board was obtained from Guangxi Guitang (Group) Co., Ltd. (Nanning, China). Silane coupling agents (KH550, KH560 and KH570) were purchased from Jinan Xingfeilong Chemical Co., Ltd. (Shandong, China). Acetyl tributyl citrate (ATBC) was purchased from Aladdin (Shanghai, China). PLA 3001D was acquired from NatureWorks LLC (Minnetonka, MN, USA). All of the other chemicals used in the study were purchased from Chongqing Chuandong Chemical (Group) Co., Ltd. (Chongqing, China) and were of analytical grade.

Preparation of MCC

Bagasse pulp was first softened using a standard pulp disintegrator (SJ0105, Intnexen Precision Instruments Co., Ltd, Guangdong, China), after which a suitable amount of the bleached pulp was mixed with distilled water for 3 min using a magnetic stirrer in a solid-to-liquor ratio of 1:30. Then, the mixture was treated with 7% HCl at 80 °C for 1 h. At the end of the reaction, the mixture was cooled to room temperature in cold water bath and neutralized using NaOH. The residue was separated by filtering and washing with distilled water and was then dried under vacuum for 24 h. The dried sample was ground into powder using a superfine pulverizer (FS30C, Jinan Feichi Instrument Co., Ltd, Shandong, China). Finally, the MCC used for subsequent modification was obtained by passing the powdered MCC through a 200 mesh sieve with a diameter of 20 cm (Adel et al. 2010).

Surface Modification of MCC

KH550 was dissolved in 90% ethanol solution (3 wt%). Next, the solution was adjusted to pH 4–5 using glacial acetic acid and prehydrolyzed at room temperature for 60 min. A constant mass of dry MCC (5 g) was put into the acidified solution (100 ml), and the mixture was shaken in an oscillator at room temperature for 2 h. The mixture was centrifuged with ethanol three times to remove residual modifier. And then the wet residue was placed in a fume hood until the alcohol solution was completely evaporated. After drying in an oven at 65 °C for 12 h, the modified MCC was stored in a sealed bag for further use.

Preparation of MCC/PLA Biocomposite Films

PLA was dissolved in dichloromethane at a solid-to-liquor ratio of 1:4 and stored overnight in a sealed container. After adding ATBC to the PLA solution (20%, mass ratio of ATBC to PLA), either unmodified MCC or modified MCC was mixed with the solution at a certain ratio and mechanically stirred for 1 h. The mixture was rested for 3 h to de-foam and treated with ultrasound until no bubbles appeared. PLA and MCC biocomposite films about 70 μm thick were obtained using an automatic film coating machine (AFA-II, Shanghai Jingyu Instrument Equipment Co., Ltd. China). The product was placed in a constant temperature and humidity incubator (25 °C and 50% humidity). Finally, the films were air-dried overnight to remove the moisture for further use.

Characterization of MCC/PLA Biocomposite Films

Mechanical Analysis

The tensile strength and elongation at breaking of MCC/PLA biocomposite films were determined using an electronic universal testing machine (Instron® Model 3367, Illinois Tool Works Inc., Norwood, MA, USA) according to the Chinese Standard GB/T 1040.3-2006. Experiments

were carried out on 150 mm long × 15 mm wide specimens at an elongation rate of 100 mm/min. The initial distance between the specimen mounts was set to 100 mm.

Surface Morphology Analysis

The microstructure of the fracture surfaces of MCC/PLA biocomposite films was analyzed using a scanning electron microscope (SU8020, Hitachi High-Technologies Corporation, Tokyo, Japan). Films were freeze-cut in liquid nitrogen, and the section for SEM analysis was retrieved using a forceps. The apparent morphology of PLA and films during degradation experiments (in soil) were observed using a digital camera (D7100, Nikon Corp., Tokyo, Japan) (Weng et al. 2013).

X-Ray Diffraction (XRD)

The crystallinity of the samples was examined using an X-ray diffractometer (MiniFlex600, Rigaku Corp., Tokyo, Japan) with a monochromatic Cu Kα radiation source in the step-scan mode with a 2θ angle ranging from 5° to 55° at a scan rate of 5°/min. The operating voltage and current were 40 kV and 30 mA, respectively. The crystallinity index (CrI) was calculated using Eq. (1):

$$\mathrm{CrI} = \frac{I_{002} - I_{\mathrm{am}}}{I_{002}} \times 100\% \quad (1)$$

where I_{002} is the maximum intensity of the diffraction from the 002 plane, and I_{am} is the intensity of scattered X-rays for the amorphous part of the sample.

Fourier Transform Infrared Spectroscopy (FTIR)

The functional groups of powdered, unmodified MCC and modified MCC were analyzed in the range of 400 cm^{-1} to 4000 cm^{-1} using FTIR (FTIR7600, Lambda Scientific Pty Ltd., Edwardstown, Australia) with a KBr pellet (Yang et al. 2017).

Thermogravimetric Analysis (TGA)

MCC samples were subjected to TGA using an integrated thermal analyzer (ZRY-2P, Shanghai Precision Scientific Instrument Co., Ltd., Shanghai, China) in the temperature range from 30 to 700 °C at 10 °C/min under a flow of nitrogen gas.

MCC/PLA Composite Film Degradation in Soil

The mass residual ratio of composite film samples buried in real soil environments was used as a parameter to evaluate the degradability of the films. The experiment was carried out at the soil pond located in the west of Guangxi, China. At the bottom of the pond wall there are two weep holes through which excess water can ooze out. The actual size of the soil pond is 4.5 m × 2.5 m × 1.5 m (length × width × height). The soil temperature, acidity level and moisture at 20 cm depth were recorded every day. The temperature, acidity level and moisture of the soil were mainly at 26 ± 3 °C, 6 ± 1 and 30 ± 5%. After being accurately weighed, biocomposite film samples 180 mm long and 20 mm wide were buried in soil at depths of approximately 0 mm, 80 mm and 200 mm. The samples were allowed to degrade in the soil for 30, 60 and 90 days. After being removed from the soil, the film samples were washed with 75% ethanol solution. The surface ethanol was washed with deionized water. Then, each clean sample was placed in an oven at 80 °C for 4 h and weighed. The mass residual ratio (P) of the sample was calculated using Eq. (2):

$$P = \frac{W2}{W1} \times 100\% \quad (2)$$

where W_1 is the mass of the sample before it was buried, and W_2 is the mass of the sample after being buried and degraded. Each test was untaken in triplicate.

Results

Effect of MCC Content on the Mechanical Properties of Biocomposite Films

To study the effect of unmodified MCC content on the mechanical properties of MCC/PLA biocomposite films, the unmodified MCC content was set to 0%, 5%, 10%, 15%, 20% and 25%. The tensile strength and elongation at breaking were measured. The amount of ATBC added was fixed at 10% (w/w).

Figure 1 shows tensile strength and elongation of tested films at breaking. Both parameters decreased as the content of unmodified MCC increased. When the added amount of unmodified MCC was 25 wt%, the elongation at breaking and the tensile strength of the films decreased by 61.53% and 55.88%, respectively, compared to 0%. The decrease occurred because the surface of the unmodified MCC contained a large amount of hydroxyl groups which is probably caused by agglomeration, resulting in poor compatibility and dispersion between the MCC and PLA (Wu and Zuo 2016, Xian et al. 2018).

PLA cannot completely enclose cellulose, which leads to problems such as surface roughness, poor mechanical properties and poor resistance to water of the biocomposite films (Kale et al. 2019). The unmodified MCC has the features of toughness, small particle size and limited ability

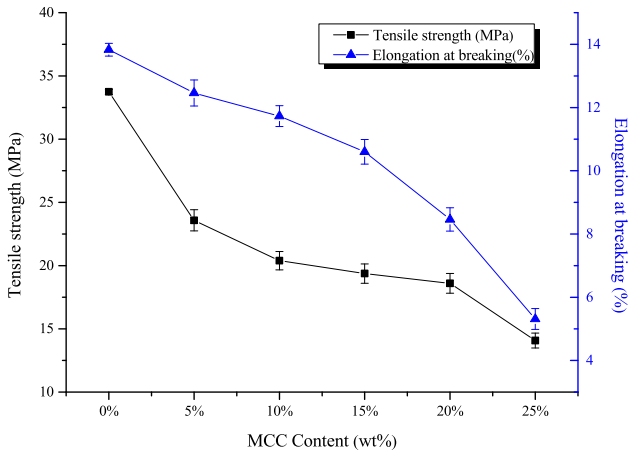

Fig. 1 Effect of unmodified microcrystalline cellulose (MCC) content (0–25%) on mechanical properties of composite films

to form an effective three-dimensional network structure in the PLA matrix (Mathew et al. 2005). Therefore, an external force exerted on the composite film made from unmodified MCC cannot be transferred to the MCC fiber (Suchaiya and Aht-ong 2012).

Characterization of Modified MCC

To increase the compatibility between MCC and PLA and improve the mechanical properties of MCC/PLA biocomposite films, MCC was modified by adding a silane coupling agent KH550 (KH550-MCC). The modified MCC was analyzed by FTIR and XRD, and the results are shown in Figs. 2 and 3, respectively.

The adsorption peaks (i.e., transmittance troughs) at 3380 cm^{-1}, 2870 cm^{-1}, 1653 cm^{-1}, 1430 cm^{-1} and 1030 cm^{-1} (Fig. 2) were due to the stretching vibration of -OH, the symmetric and asymmetric stretching vibration peaks of $-CH_2$, absorption peak of water, the symmetric

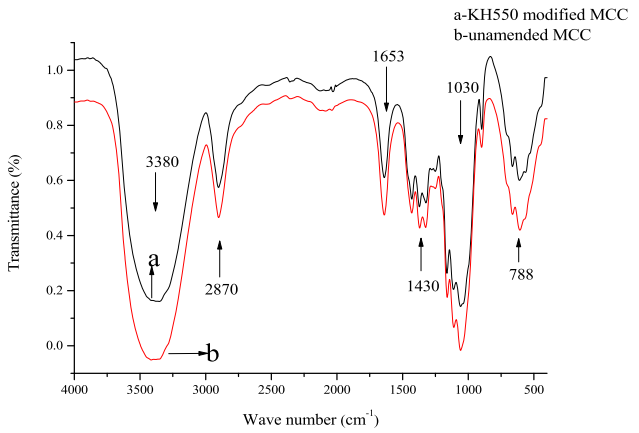

Fig. 2 Infrared spectra of microcrystalline cellulose (MCC) before and after modification with silane coupling agent (KH550)

bending vibrational peak of CH_2 in pyran ring and the stretching vibration of C–O–C, respectively. The characteristic absorption peak at 788 cm^{-1} was the out-of-plane bending vibration of the N–H bond of KH550. Some peaks were not easily observed due to the strong stretching vibration peak of cellulose at 1030 cm^{-1} and 1070 cm^{-1}; nevertheless, the intensity of the absorption peak at 1030 cm^{-1} was obviously increased after the MCC was modified with the KH550 (Chen et al. 2013).

Figure 3 shows that both unmodified MCC and KH550-MCC exhibited two main XRD peaks located at 16° and 22.5°, which corresponded to the (100) and (200) planes of the cellulose crystal lattice, respectively. These patterns indicated that the crystalline structure of MCC was not changed by the addition of KH550 (Coelho et al. 2017). Compared with the unmodified MCC, the crystallinity of KH550-MCC was reduced by 4.97% (Table 1). With the decrease in crystallinity, the decrease in the polarity of MCC helped improve the interfacial compatibility between MCC and PLA (Deshpande et al. 2000).

Fractured Surfaces Analysis of Composite Films

Figure 4a is an SEM image of the cross section of the composite films made from unmodified MCC and PLA. The unmodified MCC filled into the PLA matrix, and the cross section of the film contained a number of holes and bubbles (Lizundia et al. 2016). This pattern suggested that the compatibility between unmodified MCC and PLA was poor (Wu and Wang 2018). After the MCC was modified by the silane treatment (KH550), the surface morphology of the composite film was obviously different from that of the film made with unmodified MCC. As shown in Fig. 4b, the surface of the film made with modified MCC was almost smooth and flat, which suggested that the compatibility between MCC and PLA was improved by the silane treatment (Thakur et al. 2014).

Effect of KH550 Content on the Mechanical Properties of Biocomposite Films

To study the effect of KH550 on the tensile strength and elongation at breaking of the composite films, various amounts of KH550 (0%, 0.5%, 1%, 1.5%, 2%, 2.5%, 3% %, 3.5% and 4%, w/w) were added to powdered, unmodified MCC. The amount of ATBC added to each formulation was fixed at 10% (w/w). The results are shown in Fig. 5.

Figure 5 shows that the tensile strength and elongation at breaking of the composite films increased as the KH550 dosage increased from 0 to 3%. At the dosage of 3%, the values of both metrics were at their maximum and declined at KH550 doses exceeding 3%. When the amount of added

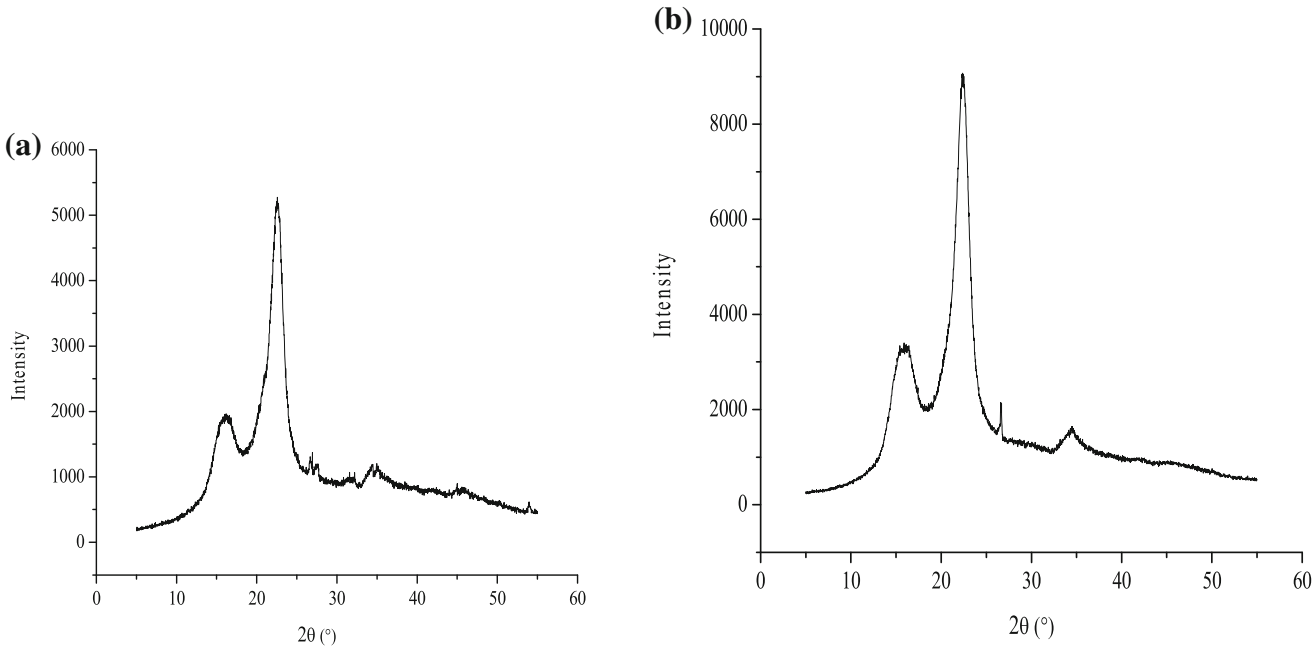

Fig. 3 X-ray diffraction patterns of **a** microcrystalline cellulose (MCC) after modification with silane coupling agent (KH550-MCC) and **b** unmodified MCC

Table 1 Crystallinity (CrI) of unmodified microcrystalline cellulose (MCC) and MCC modified with silane coupling agent (KH550)

Sample	KH550-MCC	Unmodified MCC
I_{002}	5068	9008
I_{am}	1395	2032
CrI (%)	72.47%	77.44%

I_{002} is the maximum intensity of the diffraction from the 002 plane, and I_{am} is the intensity of scattered X-rays

KH550 was inadequate, not all of the original MCC was completely modified. However, when the added amount of KH550 was excessive, the excess was hydrolyzed to silanol, and silanol oligomers were formed between the silanol groups (Wu and Wang 2018). As a consequence, the original three-dimensional network structure of the KH550 was destroyed, and a multi-molecular layer (or partially aggregated complex) was formed in the composite film system, which limited the compatibility of the MCC with the PLA matrix. When the optimum amount (3%) of KH550 was added, one end of the KH550 molecule effectively reacted with the hydroxyl group on the surface of the original MCC to form an ordered coupling agent monolayer. The hydrophobic group at the other end of the KH550 molecule was exposed, thereby improving the compatibility of MCC with PLA (Xie et al. 2010).

Effect of KH550-MCC Content on the Mechanical Properties of Biocomposite Films

The effect of KH550-MCC addition on the tensile strength and elongation at breaking of the composite films was evaluated. The KH550-MCC used in this experiment was created using the optimum amount of KH550 (3% w/w KH550 to unmodified MCC). The dosage of KH550-MCC was set at 0%, 5%, 10%, 15%, 20% and 25% (w/w modified MCC to PLA), and the content of ATBC was held constant at 10%. The results are shown in Fig. 6.

Figure 6 shows that the tensile strength and elongation at breaking of the composite film increased as the dosage of KH550-MCC increased from 0% to 20% and then decreased as doses exceeded 20%. When the dosage of KH550-MCC was 20%, the mechanical properties of the composite film reached their maximum values. Compared with the effect of unmodified MCC (Fig. 5), the effect of modified MCC on the tensile and elongation properties of composite film was obvious, increasing both metrics. After being modified by KH550, the polarity of MCC was reduced due to the decrease in the number of hydroxyl groups on the surface, so its compatibility with PLA was improved, which was consistent with the reported literature (Xiang et al. 2018). According to the data illustrated in Fig. 6, the optimum dosage of KH550-modified MCC to create MCC/PLA composite films was 20%.

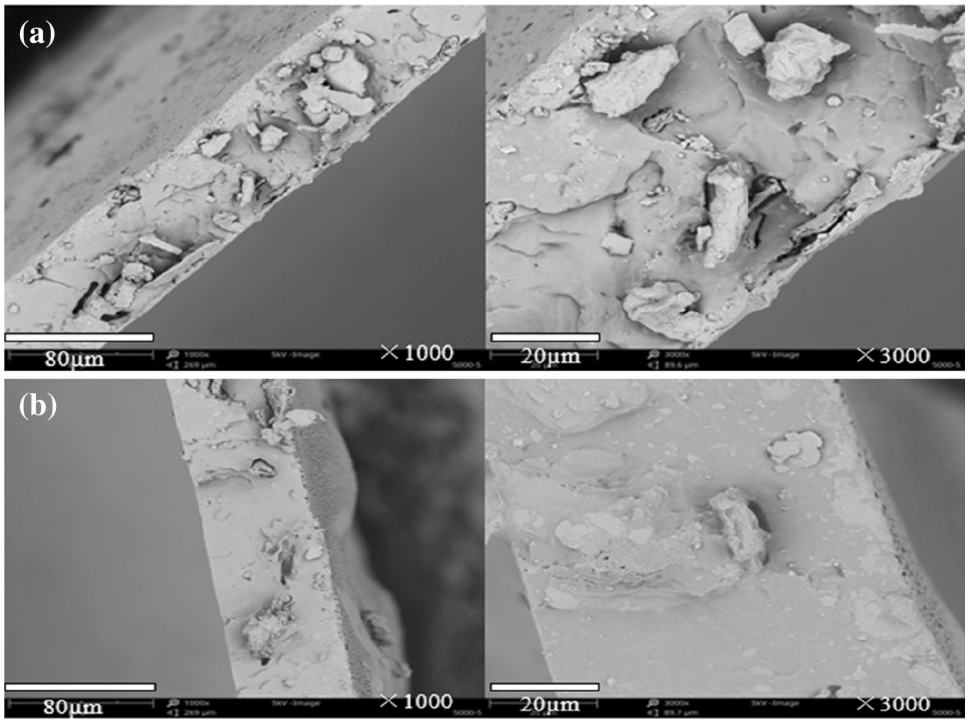

Fig. 4 Scanning electron microscope image of cross section of biocomposite films made from polylactic acid mixed with **a** unmodified microcrystalline cellulose (MCC) and **b** MCC modified with silane coupling agent (KH550-MCC)

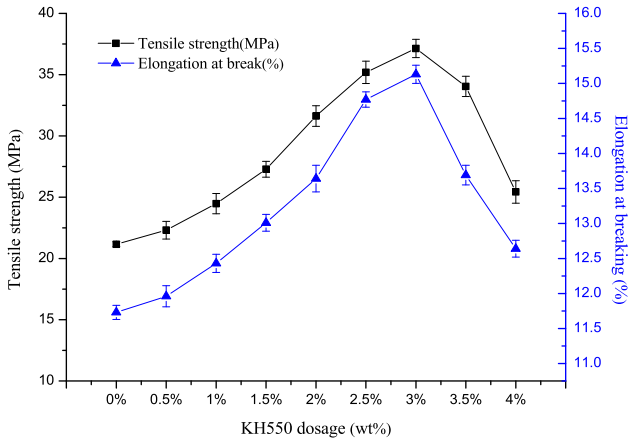

Fig. 5 Effect of silane coupling agent (KH550) dosage on the mechanical properties of composite films made from polylactic acid mixed with microcrystalline cellulose

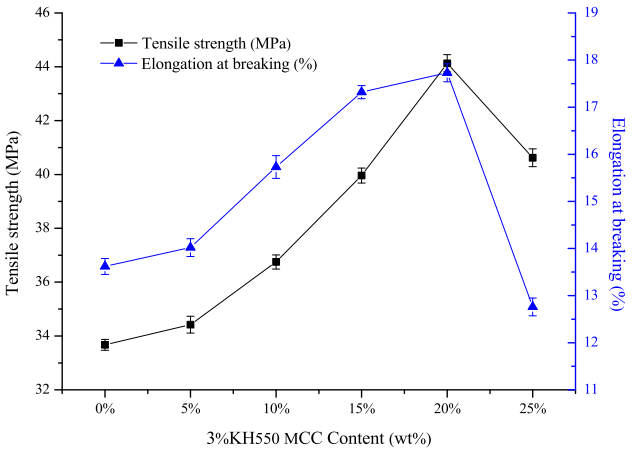

Fig. 6 Effect of modified microcrystalline cellulose (MCC, modified using 3% silane coupling agent, KH550) on the mechanical properties of composite films made from polylactic acid mixed with MCC

Effect of ATBC Content on the Mechanical Properties of Biocomposite Films

The effect of ATBC on the mechanical properties of the MCC/PLA composite films was assessed by varying the amount of ATBC added to the mixture. In the experiments 0%, 3%, 6%, 9%, 12%, 15%, 18%, 21% and 24% of ATBC were added to mixtures containing 20% modified MCC (where the unmodified MCC was modified by 3% KH550 w/w). The results are shown in Fig. 7.

Figure 7 shows that as the dosage of ATBC increased, the tensile strength of the composite films gradually decreased, while the elongation at breaking increased. The elongation at breaking increased obviously at dosages of ATBC exceeding 12%. This occurred because the plasticizing effect of ATBC was exerted only on PLA (Arrieta et al. 2016). When the dosage of ATBC was less than 12%, the interaction between the PLA molecules was greater than the interfacial bonding strength between KH550-MCC and PLA. Consequently, the fracture of the composite films was mainly due to brittle fracturing between MCC and PLA molecules.

When the dosage of ATBC exceeded 12%, the interfacial bonding strength between KH550-MCC and PLA was

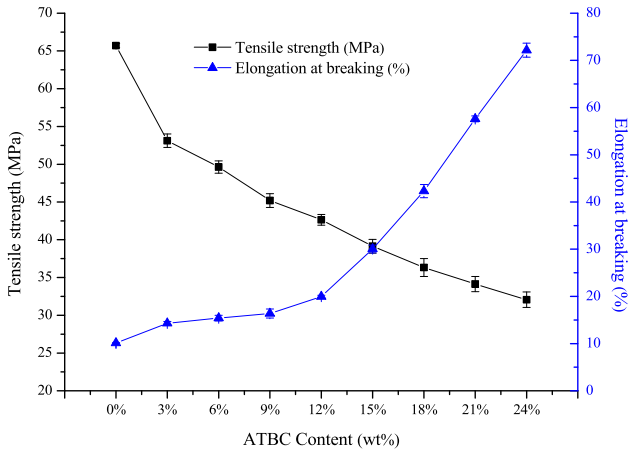

Fig. 7 The effect of acetyl tributyl citrate (ATBC) content on the mechanic properties of composite film made from polylactic acid mixed with microcrystalline cellulose

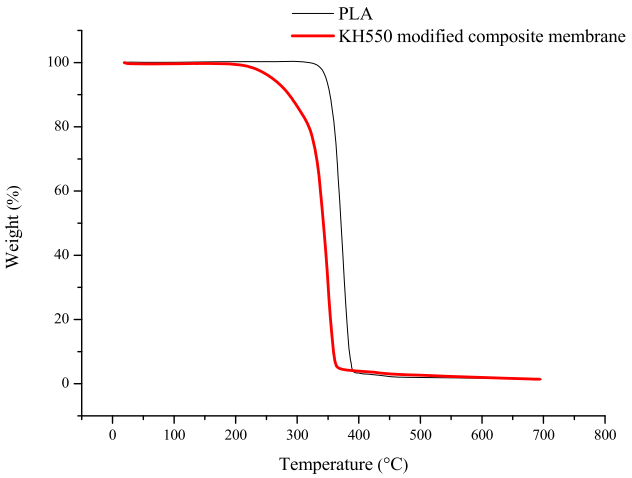

Fig. 8 Thermal weight curves of films made from **a** polylactic acid (PLA) only and **b** PLA mixed with microcrystalline cellulose (MCC) modified with silane coupling agent (KH550)

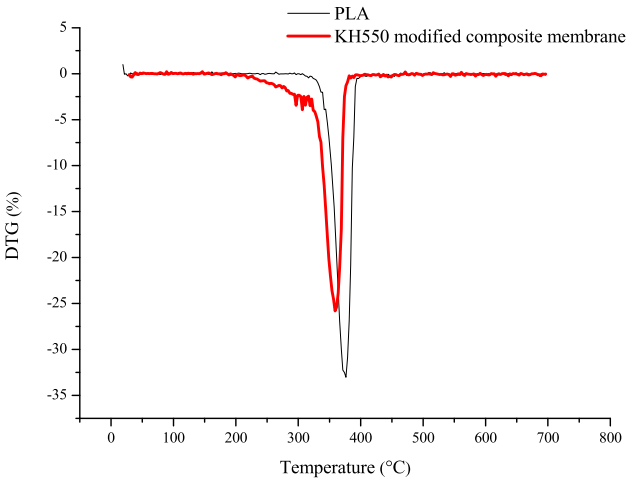

Fig. 9 Derivative thermogravimetric (DTG) curves of film made from **a** polylactic acid (PLA) only and **b** PLA mixed with microcrystalline cellulose (MCC) modified with silane coupling agent (KH550)

greater than the interaction between the PLA molecules. Consequently, when an external force was applied, the composite film experienced deformation yield (rather than fracturing) between the PLA molecules. Therefore, at ATBC doses exceeding 12%, the elongation at breaking of the composite film sharply increased. When the dosage of ATBC was 15%, the elongation at breaking of the composite film increased by 296.74% compared to that at ATBC dosage of 0%, but the tensile strength at comparable ATBC doses decreased by 40.42%. Based on the data illustrated in Fig. 7, an ATBC dosage of 15% was selected as the optimal ratio for making modified MCC/PLA composite films.

Thermogravimetric Properties

The thermal stability of film made from only PLA was compared to that for KH550-MCC/PLA composite film. The thermogravimetric curves and the derivative thermogravimetric curves for the two types of film are shown in Figs. 8 and 9, respectively.

Data illustrated in Figs. 8 and 9 show that the PLA film experienced a significant weight loss at 344.2 °C. However, when the temperature advanced to 390.5 °C, no change of mass occurred. The overall mass reduction was 1.75%. In contrast, the KH550-MCC/PLA composite film experienced weight loss at a much lower temperature (246.4 °C). When the temperature advanced to 363.9 °C, no change in mass occurred, and the final mass reduction was 2.03%. By comparison, the overall mass losses of the two types of films were not very different, but the initial decomposition temperature of the KH550-MCC/PLA composite film was much lower and the weight loss rate was slower than that of the PLA film. This analysis showed that when KH550-MCC was used as a "filler" to reduce the thermal stability of the composite films (by reducing the force between the polymer molecules) (Wu and Wang 2018), the higher thermodynamic properties of the PLA matrix were substantially unaffected.

Effect of KH550 on the Degradation of Unburied and Buried Biocomposite Films

The effect of modified MCC on the degradation in soil of PLA composite films was assessed by comparing the degradation of films made using (a) PLA alone, (b) unmodified MCC and PLA and (c) KH550-MCC and PLA. In this experiment, the film thickness and the burial depth were 73 μm and 8 cm, respectively. The results are shown in Fig. 10.

Figure 10 shows that the mass of buried PLA film samples decreased only slightly (≈ 5%) after 90 days. After 90 days, the degradation of the films (as indicated by changes in mass) from small to large followed the order PLA, KH550-MCC/PLA and MCC/PLA. Once PLA was hydrolyzed into small molecules, it was decomposed in the soil by microorganisms. However, because PLA is a hydrophobic resin, it resists being hydrolyzed by soil moisture (Weng et al. 2013). Consequently, the degradation of PLA film was the slowest of the three films tested.

Compared with PLA films, the MCC/PLA and KH550-MCC/PLA composite films degraded more easily, apparently due to the addition of MCC (Vasile et al. 2018). Because MCC adsorbs water, degradation of the PLA in the composite films by water absorption was promoted (Kale et al. 2019). However, as indicated in Fig. 10 the degradability of KH550-MCC/PLA composite film was significantly lower than that of MCC/PLA film. This difference was explained by noting that KH550-MCC had much better compatibility with PLA matrix than did unmodified MCC; therefore, the KH550-MCC actually hindered the hydrolytic degradation of the composite film. In addition, the polarity of the surface of MCC was reduced by KH550, making water absorption degradation of PLA more difficult.

Visual Changes in Films After Burial

The degradation in soil of the films also was assessed visually by comparing the apparent morphological changes of films made using (a) PLA alone, (b) unmodified MCC and PLA and (c) KH550-MCC and PLA (Fig. 11). The appearance of the films was recorded using a digital camera at 0, 30, 60 and 90 days following burial. All films were buried 8 cm deep.

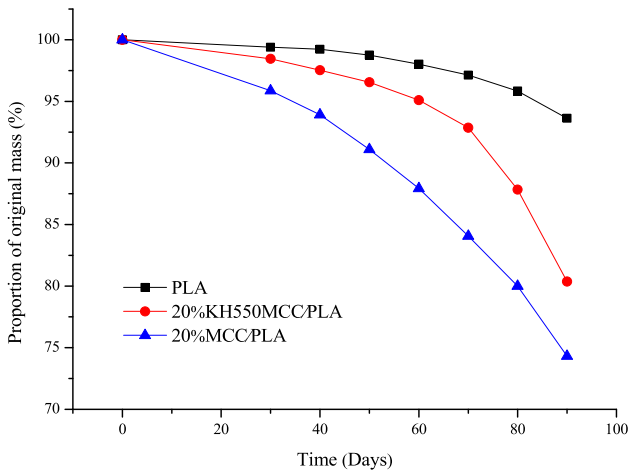

Fig. 10 Degradation in soil of films made using polylactic acid (PLA) alone, 20% unmodified microcrystalline cellulose (MCC) combined with PLA and 20% MCC modified using silane coupling agent (KH550) combined with PLA

Based on visual appearance, the order of biodegradation was MCC/PLA > KH550-MCC/PLA > PLA. The surface of PLA film exhibited no change within 30 days, but afterward showed some fine marks accompanied with surface wrinkles. After 60 days, the wrinkling of the PLA film was more obvious, and some very obvious white points appeared. Furthermore, the transparency of the PLA film gradually decreased as the degradation time increased. The hydrolysis of low molecular weight branches in the PLA may have been responsible for the loss of transparency (Nazhad et al. 2015).

Compared with PLA film, composite film made from PLA combined with unmodified MCC exhibited a yellow area on the surface, and the unevenness on the film surface was accompanied by a small amount of material shedding. As described in Section "Effect of KH550 on the degradation of unburied and buried biocomposite films," hydrolysis of PLA and the degradation of the composite membrane were improved by adding MCC. As noted previously, the surface of MCC is highly hydrophilic and the material can adsorb water from the soil, thereby advancing water absorption degradation of the PLA. In addition, the compatibility between the unmodified MCC and the PLA matrix was poor and there were more cavities and voids at the interface; thus, the hydrolysis process of the PLA around the MCC was accelerated. The yellowing of the surface may have been caused by the metabolism of soil microorganisms.

Compared with the MCC/PLA film, the KH550-MCC/PLA composite film exhibited surficial changes later. Due to the addition of KH550, the compatibility of MCC with the matrix PLA was improved and the water absorption of the MCC was reduced. Therefore, enrichment of water degradation by MCC and the promotion of hydrolysis of PLA were reduced.

Conclusion

In this study, tensile strength and elongation at breaking were used to determine the optimum conditions for the preparation of composite films containing PLA, MCC, KH550 and ATBC. The optimum formulation was 3% KH550 (to modify the MCC), 20% KH550-MCC and 15% ATBC. KH550-modified MCC improved the interfacial compatibility of KH550-MCC/PLA composite films, obviously increasing the tensile strength and elongation at breaking, and making the film more easily degraded when heated. The soil burial experiment showed that the degradation rate of the composite films was increased by adding MCC. During the soil degradation process, the

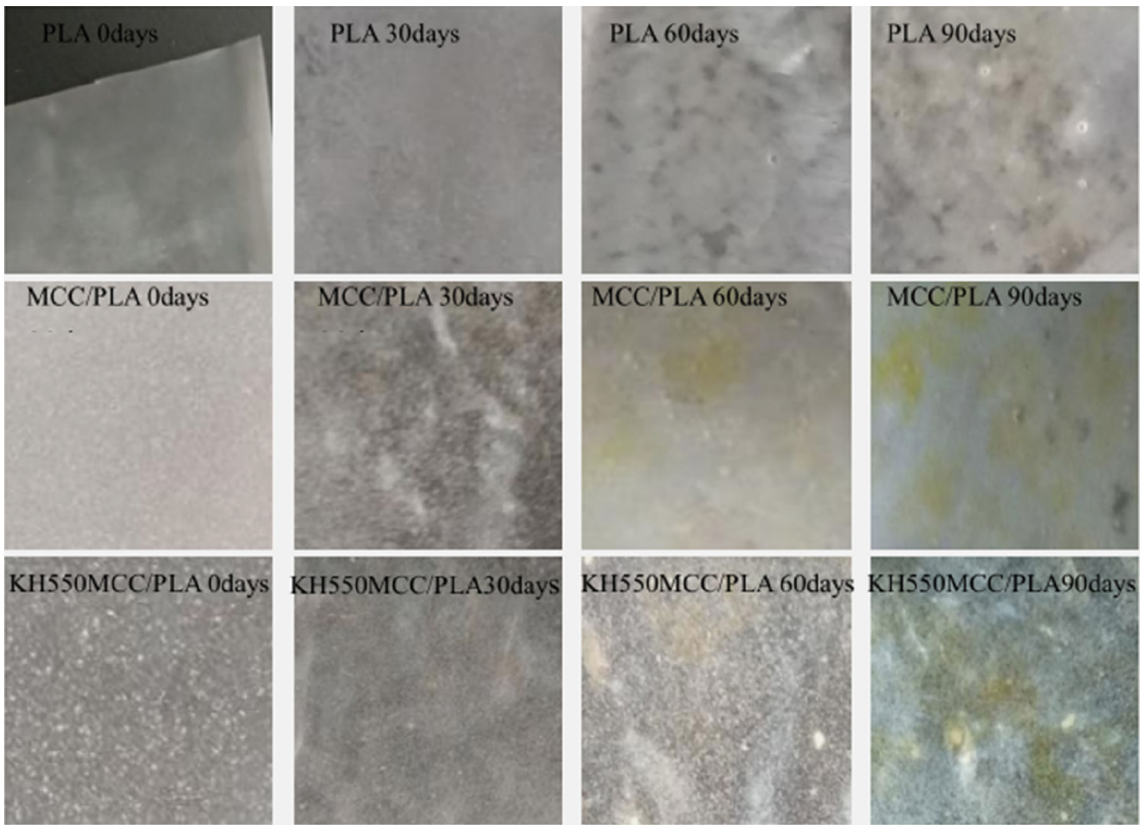

Fig. 11 Apparent morphological changes during degradation in soil of films made using polylactic acid (PLA) alone, unmodified microcrystalline cellulose (MCC) combined with PLA (MCC/PLA) and MCC modified using silane coupling agent mixed with PLA (KH550-MCC/PLA)

transparency of the composite films was reduced, the color became yellow, and the surface wrinkling was obvious. Because the compatibility between the MCC and the PLA matrix was improved by KH550, the degradation of the KH550-MCC/PLA composite film was ranked between that of film made from PLA alone and that of composite film made from unmodified MCC and PLA.

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