



Carboxylated styrene-butadiene rubber adhesion for biopolymer product-based from cassava starch and sugarcane leaves fiber

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ABSTRACT

The biopolymer-based from cassava starch (CS) and cellulose fiber (CF) from sugar cane leaves was prepared by help of carboxylated styrene-butadiene rubber (CSBR) as an adhesion. The CF was isolated by using acid hydrolysis under heating (50 wt%, 140 °C for 2 h). The size, morphology, chemical structure, interaction bonding, dynamic mechanical behavior, thermal analysis, biodegradation, physical and mechanical performance of CSBR/CS/CF composites were investigated. The highest tensile value was found at 1.6 wt% CF loading (27.96 MPa) while preserving the large extent of their biodegradability (< 80% after buried in soil for 30 days). Moreover, the increasing CF content led to decrease in the water contact angle, moisture content owing to the interfacial adhesion between carboxyl groups on the CSBR and hydroxyl groups on the CF surfaces. This study demonstrated a significant concentration of the CF on film properties. Moreover, these inexpensive biodegradable composites were preferable for packing, fruit container and paper coating applications.

1. Introduction

Cassava (*Manihot esculenta* Crantz) is one of the economic crops in Thailand. Nowadays, it is used as raw material in varied industries, such as food, animal feed, adhesion component, paper, and bioethanol. In addition, CS has been extensively studied to produce biodegradable films (Mohsenabadi et al., 2018) by many processing techniques, such as casting (Condés et al., 2018), injection (Wang, 2014) and blow molding (Wang et al., 2017). Because it is able to form transparent film, good film-forming, excellent thickening character, low gelatinization temperature, good gel stability, and semi-permeable to carbon dioxide-oxygen (Stagner et al., 2012). It has been stated that the CS is promising candidates for future materials (Li and Cho, 2017). However, the CS based films are limited in their applications due to its poor mechanical strength, high water sensitivity and brittle character compared to synthetic materials (Bher et al., 2018). A solution to these problems, a native starch was modified by using various methods (Goudarzi and Shahabi-Ghahfarrokhi, 2018; Ma et al., 2017; Liu, et al., 2016a,b) to obtain high-performance and desirable properties. For example, chemical modifications have been reported, such as graft copolymerization (Haroon et al., 2018), crosslinking (Kou and Gao, 2018), and acetylation (Sizing Bismark et al., 2018) for introducing a new substituent group to starch molecule leading to good required properties. Another method is blending of two or more different polymers with/without any

chemical bonding between them to achieve various property combinations of the resulting film (Muller et al., 2017).

Previous study reported compounding the styrene-butadiene rubber (SBR) with starch for improving the strength and hydrophobic character (Wu et al., 2006). Furthermore, carboxylated styrene butadiene rubber (CSBR) which is a terpolymer of styrene, butadiene, and small amount unsaturated carboxylic acid was employed to improve adhesion between them. Riyajan reported the biocomposite made from the CS and CSBR. The 1/1 CS/CSBR blend exhibited the highest tensile strength (25 MPa) and high adhesion between them. The maximum biodegradation and maximum swelling ratio (200%) was found in the 1/1 CS/CSBR which increased as a function of CS portion in the blend. In addition, the composite hardly decomposed in environment owing to a high rubber portion (Riyajan, 2018).

Nowadays, cellulose from plant was added in starch-based materials to enhance the mechanical strength instead of inorganic fillers owing to a high energy consumption and environmental pollution (Zhang et al., 2014). Pelissari et al. reported adding cellulose nanofibers from banana peel in starch matrix. Results showed the homogeneous dispersion of cellulose in the matrix resulting in increasing the glass transition temperature owing to the strong interactions between them (Pelissari et al., 2017). Bras et al. reported blending cellulose whisker from sugarcane bagasse pulp in natural rubber matrix. Results showed the existence of cellulose whiskers enhanced the rate of degradation and its value was

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62% weight loss after buried in soil for 4 weeks at 7.5 wt% cellulose whiskers loading (Bras et al., 2010). Cellulose nanocrystals (CNs) showed a significant reinforcing effect on the CSBR latex (Cao et al., 2013). Results revealed the uniform dispersion in the CSBR latex, improved tensile value (24.1 MPa) and tear strength (65.2 MPa) with loading CNs up to 15 phr. Other work, sisal fiber was added in starch to form a new polymer that is promising packed product replacement for expandable polystyrene (Xie et al., 2018). Sisal fiber content affected the cushioning property by changing the density of the structure. The composites which are better than the expandable polystyrene were compressed under 0.7 to 6 MPa loading. In this work, we attempted to modify a native CS by using a small proportion of CSBR latex in the CSBR/CS blend (0.1/1) to preserve the large extent of their biodegradability and employing cellulose fiber (CF) from sugarcane leaves to improve the mechanical strength. To the best of our knowledge, there is no study adding the CF in the CSBR/CS matrix by help of the CSBR latex as an adhesive. The CSBR/CS/CF composites were prepared by casting method. Moreover, the effects of the CF on the mechanical properties, moisture content, swelling ratio, surface wettability, physical and thermal properties of these composites were investigated in this work. After used, the biodegradation of the composites was also evaluated.

2. Materials and methods

2.1. Materials

The CSBR latex emulsion (Rovene® 9410) was obtained from Mallard Creek Polymers (Charlotte, North Carolina, USA). The styrene/butadiene ratio is 25/75. The glass transition temperature and solid content are -56 °C and 54% respectively with pH ~8.5. The CS containing 32% amylose was derived from General Starch Ltd. (Bangkok, Thailand). The potassium persulfate ($K_2S_2O_8$, Assay ≥ 98%) was supplied from Fluka, Seelze, Germany. 10% Triton™ X-100 as a non-ionic surfactant and α -cellulose were purchased from Sigma-Aldrich. Glycerol ($C_3H_8O_3$, Assay ≥ 99.5%) as plasticizer and sodium hydroxide anhydrous pellets (NaOH, ACS reagent) were supplied from CARLO ERBA Reagents. Sulfuric acid (H_2SO_4 , AR. grade 98%) was obtained from RCI Labscan, Thailand.

2.2. Extraction cellulose fiber from wasted sugarcane leaves

The procedure of preparing cellulose fiber was modified with according to previous work (Liu, et al., 2016a,b). 200 g Wasted sugarcane leaves were cut into small pieces, bleached in 500 g 10% w/w sodium hydroxide solution, washed with deionize water to neutral pH, and dried in an oven. The acid hydrolysis was performed using sulfuric acid (50 wt%) and the treated sugarcane leaves (50 g) was further treated with volume concentrations of 10 g 50% sulfuric acid taken in 10:1 ratio under vigorous stirring at 140 °C for 7 h. Subsequently, the sediment was diluted 10-fold with deionized water to stop hydrolysis reaction follow by centrifugation (10000 rpm, 15 °C, 15 min), and then dialyzed against deionized water until the pH was neutral. Finally, the suspension was carried out through ultrasonic treatment (15 min), which then was stored in a refrigerator.

2.3. Preparation of the CSBR/CS/CF composites

CS (50 g) was dispersed in 950 mL of distilled water at 90 ± 3 °C under stirring resulting in the CS paste. Potassium persulfate (KPS) solution (3 g in aqueous solution) was added to the mixture. KPS is used to improve chemical interaction between the CSBR and CS. CSBR latex (22 g) was homogenized with the gelatinized CS at ratio 0.1/1 in the presence of Triton™ X-100 (1 g in DI water 10 mL) at 220 rpm, 70 °C for 1 h. The 0.1/1 CSBR/CS ratio was chosen for this work due to easy biodegradation in natural work. The CSBR/CS blend was obtained and used in further step. The 0.1/1 CSBR/CS blend was mixed with CS gel

(CS 50 g in DI water 500 mL) at ratio 1/1 which is an optimal condition before separation between the CS and CSBR get a mixture suspension. Subsequent, glycerol (2 wt%) and CF by changing the CF contents over the range of 0, 0.4, 0.8, 1.2, 1.6, and 2 wt% were added to the mixture suspension. Finally, the mixture suspension was cast on glass plate (15 cm × 15 cm) and then left at 28 °C for 96 h.

2.4. Characterizations

The effects of the CF on the physical and mechanical properties of composites were studied. Size distribution and zeta potential of the CF were measured by particle size analyzer (LA-950V2 HORIBA; Japan). Contact angle measurement (TL100 Biolin Scientific; Finland) was used to determine wettability and hydrophilic behavior of samples by the sessile drop method at room temperature. An apparent contact angle (θ) was recorded in 10 s with the OneAttention software. The swelling power of the CSBR/CS/CF composites was determined. Samples were weighted before being immersed in 25 mL of deionized water at room temperature for 3 h. Subsequently, the samples were lightly wiped the excess water and immediately weighted in accordance with ASTM D570-98. The swelling ratio was determined by the following Eq. (1):

$$\text{Swelling ratio (\%)} = (W_s - W_d) / W_d \times 100 \quad (1)$$

Where, W_d and W_s are the weight of the samples before and after immersion in water (g), respectively. An average value of the three replicates for each specimen was reported. Moisture content was measured as the loss of the sample after drying it in an oven at 110 °C, calculated by Eq. (2), where W_i and W_f are the weights before and after the drying process:

$$\text{Moisture content (\%)} = (W_i - W_f) / W_i \times 100 \quad (2)$$

Moisture absorption with ASTM D570 – 98(2018) was calculated based on the weight measurement before and after moisture treatment. Samples were cut into $1 \times 1 \text{ cm}^2$ size and dried at 120 °C for 30 min. The dried samples were placed in a desiccator containing copper sulfate (RH ~ 98%). The weight of samples was measured every specific time until reaching a constant weight. The moisture absorption was calculated as follows (3):

$$\text{Moisture absorption (\%)} = (W_t - W_0) / W_0 \times 100 \quad (3)$$

Where, W_0 is the weight of dried sample and W_t is the weight gain at time t . An average film thickness was measured using the dial thickness gauge (SM-112, Teclock; Japan) in a random 5 positions. The chemical structure and interaction bonding were characterized by IR spectrometer (Spectrum 100 PerkinElmer; USA) equipped with an attenuated total reflectance device for solid analysis. The analysis performed in the range of 4000 to 500 cm^{-1} with 64 scans recorded at a 4 cm^{-1} resolution. XRD diffractometer (X'Pert MPD PHILIPS; Netherlands) was used to study the crystalline structure. JSM 7800 F (JEOL, Japan) scanning electron microscopy was used to observe the surface and cryogenically fractured surfaces which were sputter-coated with gold prior observation. The tensile tests were determined with dumbbell specimens according to ASTM D638 (6 mm width in cross section) by using a Universal Testing Machine (Instron Model 2701) with a crosshead speed of 100 mm/min. The type of specimen that has been used in type V. Dynamic mechanical analyser (DMA1 METTLER TOLEDO; Switzerland), working in tensile mode, has been used to determine the dynamic mechanical behaviors of CSBR/CS/CF composites. The temperature was run from -50 to +250 °C using a heating rate 5 °C/min at a constant frequency of 10 Hz. The samples were approximately cut $10 \text{ mm} \times 1 \text{ mm} \times 6 \text{ mm}$ (length × thickness × width). A thermogravimetric analyser (TGA/DSC3+/HT/1600 METTLER TOLEDO; Switzerland) was used to identify the thermal stability of samples by measuring the weight loss under nitrogen atmosphere. The samples were run from 30 to 850 °C at a heating rate of 10 °C/min. Biodegradation of the CSBR/

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