

Development and Characterization of Polyvinyl Alcohol/Bacterial Cellulose Composite for Environmentally Friendly Film

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ABSTRACT

This research aimed to develop and characterize polyvinyl alcohol (PVA)/bacterial cellulose (BC) composite for environmentally friendly films. BC was produced from a high-performance strain of *Komagataeibacter intermedius* BE073 isolated from a bio-extract sample. The film was prepared by varying the ratio between PVA and BC content, and treatments consisted of 100:0, 90:10, 80:20, 70:30, and 60:40. The characterization of PVA/BC film in terms of mechanical properties, film structure, water and oxygen resistances, thermal stability, and biodegradation were investigated. Results revealed that PVA/BC film had properties superior to that of pure PVA film, and it has a high biodegradation rate. The mechanical properties changed little with the addition of BC, but the tensile strength and Young's modulus increased with the addition of BC. Water absorption and moisture content were also reduced. However, adding BC improved oxygen transmission rates and thermal stability properties. Most importantly, the addition of BC helped the film to degrade. The higher the amount added, the higher the natural decomposition rate.

Keywords: Polyvinyl alcohol, Bacterial cellulose, Biodegradable film, *Komagataeibacter intermedius* BE073.

INTRODUCTION

At present, films used in different fields, such as food and agriculture, are usually made of synthetic plastics, such as polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC). These plastics degrade difficultly after use, and it takes a long time to be destroyed at a landfill (Mousa et al., 2016). As a result, leaving chemical residues is detrimental to the ecosystem and causes environmental pollution. However, at present, biodegradable films have been developed, including polylactic acid (PLA) (Mathew et al., 2005; Graupner et al., 2009), polycaprolactone (PCL) (Siqueira et al., 2009; Zhao et al., 2010), and polyvinyl alcohol (PVA) (Chakraborty et al., 2006; Chen et al., 2007; Zhang et al., 2011; Senna et al., 2014). PVA is a polymer commonly used in film production and popularly applied in

the medical field (Jiang et al., 2019) and in agriculture because it can decompose with no toxicity and is water soluble (Grunlan et al., 2004; Abdulkhali et al., 2013). However, it has been found that the mechanical properties of PVA are incomplete (Santi et al., 2019). It is also necessary to mix in additives, such as starch (Zhai et al., 2002; Jayasekara et al., 2004) and sodium alginate (Yeom et al., 1998; Adoor et al., 2007), but the additives are often costly and may require high dosages. However, studies have found that when adding xylan (Wang et al., 2014) and cellulose (Niu et al., 2015; Abdel Bary et al., 2018) to PVA, the ingredients respond to improve the properties of PVA. Unfortunately, adding xylan or cellulose from plants requires removing other components and complex material preparation processes.

Cellulose products contain another class of cellulose produced by microorganisms.

Specifically, certain bacteria can produce acetic acid (acetic acid bacteria) and cellulose fibers, called bacterial cellulose (BC). Considering the properties of this cellulose fiber, the BC has higher purity than plant cellulose because of free lignin, hemicellulose, and another compound. BC has many good characteristics, such as biodegradability and biocompatibility (Iguchi et al., 2000), high absorption, high tensile strength, high water binding capacity, and highly porous structure (Backdahl et al., 2006; Hong and Qiu, 2008; Dahman, 2009). Therefore, it has been used widely in packaging products, biomedical materials, and biofilm (Alvarez et al., 2004; Czaja et al., 2006).

Therefore, this research aimed to use BC as an additive in combination with PVA to develop a biodegradable film. This will help to improve the properties of PVA film and study the properties of the obtained PVA/BC film.

MATERIAL AND METHODS

Production of BC raw material

Komagataeibacter intermedius BE073 was used as the producer strain for BC production. This strain was identified from a bio-extract sample produced by a farmer group in Nakhon Nayok Province, Thailand (Singhaboot and Kroeksakul, 2022). *K. intermedius* BE073 was transferred to a Hestrin–Schramm medium (HS medium) for inoculum preparation (Hestrin and Schramm, 1954). The HS medium contained (g/L) 20 g of glucose (Scharlau, Spain), 5 g of peptone (Scharlau, Spain), 5 g of yeast extract (Scharlau, Spain), 2.7 g of disodium hydrogen phosphate (Na_2HPO_4) (QRëC, New Zealand), and 1.15 g of citric acid

(Scharlau, Spain). The pH of the medium was adjusted to 4.2 and sterilized by autoclave at 121 °C for 15 min. The culture was incubated at 30 °C for 3 days. After that, 10% (v/v) of the culture was inoculated into a fresh HS medium and incubated at room temperature for 14 days. Finally, the BC film was collected for raw material in the production of films.

Preparation of BC for raw material in film production

The BC film was washed and soaked thoroughly with distilled water for two to three days. It was then boiled in 1 M sodium hydroxide solution (NaOH) (Scharlau, Spain) for 1 hour and washed with distilled water until reaching a neutral pH. Then, the purified BC sheet was obtained as shown in Figure 1a. After that, the BC film was dried by freeze dryer and ground by ultra centrifugal mill (Ultra centrifugal mill ZM 200, Retsch, Germany) and a BC particle size of 180 µm was obtained (Figure 1b).

Preparation of PVA/BC biodegradable films

PVA/BC films were prepared by a solvent casting method. PVA solution was prepared by dissolving 9 g of PVA (Tokyo Chemical Industry Co., Ltd., Japan) and 1.5 g of glycerol (Ajax Finechem, Australia) in 100 mL of distilled water under stirring at 90 °C for 1 hr. A suitable BC solution was studied, and the film blended with PVA solution was prepared by introducing the dosage of BC 0.8 g was added and dissolved in 100 mL solution of 1% (w/v) potassium hydroxide (KOH) (Carlo Erba, Italy), 2% (v/v) hydrogen peroxide (H_2O_2) (Sigma-Aldrich, Germany), and 1% (v/v)

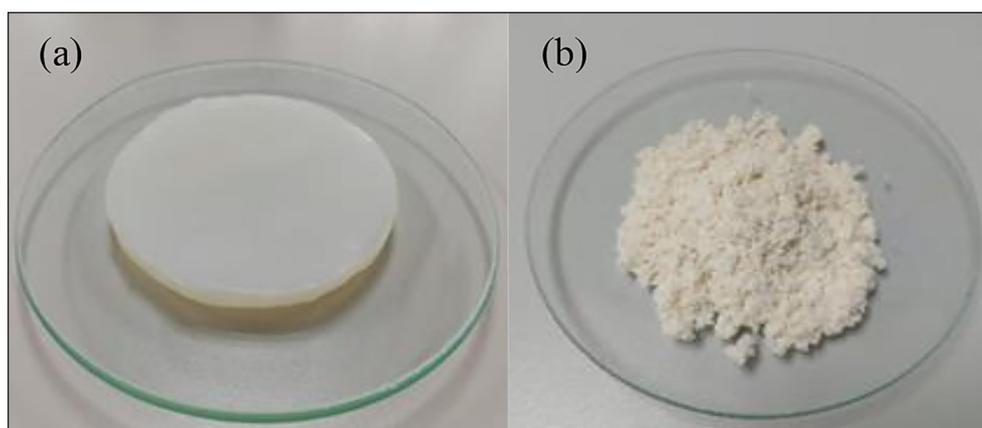


Figure 1. BC sheet after purification (a) and BC particle size of 180 µm was obtained from grinding (b)

Table 1. The composition of PVA and BC solution for film preparation

Solution	PVA solution/100 mL		BC solution/100 mL			
	PVA (g)	Glycerol (g)	BC (g)	KOH (%)	H ₂ O ₂ (%)	Nitric acid (%)
KOH	9.0	1.5	0.8	1.0	-	-
H ₂ O ₂	9.0	1.5	0.8	-	2.0	-
H ₂ O ₂ + nitric acid	9.0	1.5	0.8	-	2.0	1.0

nitric acid (Ajax Finechem, Australia), as shown in Table 1. The mixture was stirred for 2 hours at 90 °C. The mixture was allowed to cool until ambient temperature, and the pH was adjusted to neutral. After that, the PVA and BC solution were mixed at a 1:1.25 ratio until homogeneous suspension. Subsequently, the PVA/BC suspension was stirred, cast on a glass plate, and dried at 50 °C for 24 hours. In a further step, determining the best BC preparation solution involved investigating the optimum ratio between PVA and BC content for the preparation of composite film.

Characterization of PVA/BC films

Film thickness

The film thickness was measured by a micrometer vernier caliper at three different places of the PVA/BC film. Then, the value of the three measurements was averaged.

Structure analysis

The PVA/BC film structure was analyzed using a Fourier transform infrared spectrophotometer (FTIR) (Nicolet iS5 FTIR Spectrometer, Thermo Fisher Scientific, United States) and scanned in the spectral range between 4000 – 400 cm⁻¹ at room temperature.

Water absorption

The triple PVA/BC film (2×2 cm²) sample in each treatment was dried in a hot air oven at 60 °C for 24 hrs. Then, the sample was weighed (W_i) and soaked in 50 mL of distilled water. After 24 hrs, the final weight was measured (W_f), and water absorption was calculated according to the following equation (Sonker et al., 2018):

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

Moisture content

The moisture content of the BC films was calculated based on their water loss after drying. The dehydrated BC film was weighed to obtain an initial weight of the sample (W_w). After that, the sample was dried at 60 °C until reaching a constant weight and weighed for the dry weight (W_d). The water content was calculated according to the following equation:

$$\text{Moisture content (\%)} = \frac{(W_w - W_d)}{W_w} \times 100 \quad (2)$$

Oxygen transmission rates (OTR) and water vapor transmission rates (WVTR)

A test for the OTR of the film was carried out according to the ASTM D3985 method (ASTM, 2002). A test range of film between 0.005 – 2000 cc/(m²·day) was used for analysis by oxygen transmission rate analyzer (OX-TRAN 2/22, MOCON Inc., United States). The temperature and relative humidity were controlled at 23 °C and 0%, respectively. The WVTR of the film was investigated using the water vapor transmission rate analyzer (PERMATRAN-W 3/34, MOCON Inc., United States). The machine measured 0.005 – 1000 g/(m²·day) by following the ASTM F1249 method (ASTM, 2013). The OTR and WVTR were determined from the slope of the regression analysis of weight loss and time.

Mechanical properties test

The mechanical properties of PVA/BC films, including the tensile strength, elongation at break, and Young's modulus, were determined with a universal testing machine (Instron 5966, Instron, United States) according to the ASTM D882 method (ASTM, 2002). Five samples in each treatment (100 ×10 mm) were investigated at an initial gauge length of 50 mm, equipped with a 1 kN load cell and a 10 mm/min crosshead speed.

PVA/BC film surface morphology characterization

The surface morphology of PVA/BC films was recorded using a scanning electron microscope (JSM-6610, JEOL Ltd., Japan). The film was coated with gold before examination and examined at an accelerating voltage of 15 kV.

Thermogravimetric analysis

The film samples were thermogravimetrically analyzed using a thermogravimetric analyzer (TGA) (TGA 550, TA Instruments, United States). It was dried and analyzed in a temperature range of 50 °C – 600 °C. The heating rate of 10 °C/min and under a nitrogen atmosphere of 50 mL/min were used for analysis. The weight of each sample was approximately 10 mg.

Biodegradation test

Biodegradation was investigated with the soil burial degradation method (Thakore et al., 2001). The initial weight of samples (3×3 cm²) was

evaluated (W_t) and recorded, and subsequently, the samples were buried at a soil depth of 5 cm under the surface. After a test time of 7 and 14 days, samples were collected and removed from the soil. The dried weight of the films was determined (W_0), and the biodegradation rate was calculated based on the weight loss using the following equation:

$$\text{Weight loss (\%)} = \frac{(W_t - W_0)}{W_t} \times 100 \quad (3)$$

RESULTS AND DISCUSSION

Effect of BC solution on PVA/BC film

Effect of BC solution on PVA/BC film properties according to experimental Figure 2. The mechanical properties of the thin films in each solution are different when preparing BC in a KOH solution, PVA/BC thin films have the highest Young's modulus (deformability, such as bending ability) of 168.18 ± 25.29 MPa, compared to other solutions. For the BC prepared in a 2% H₂O₂ solution, it was found that the maximum tensile

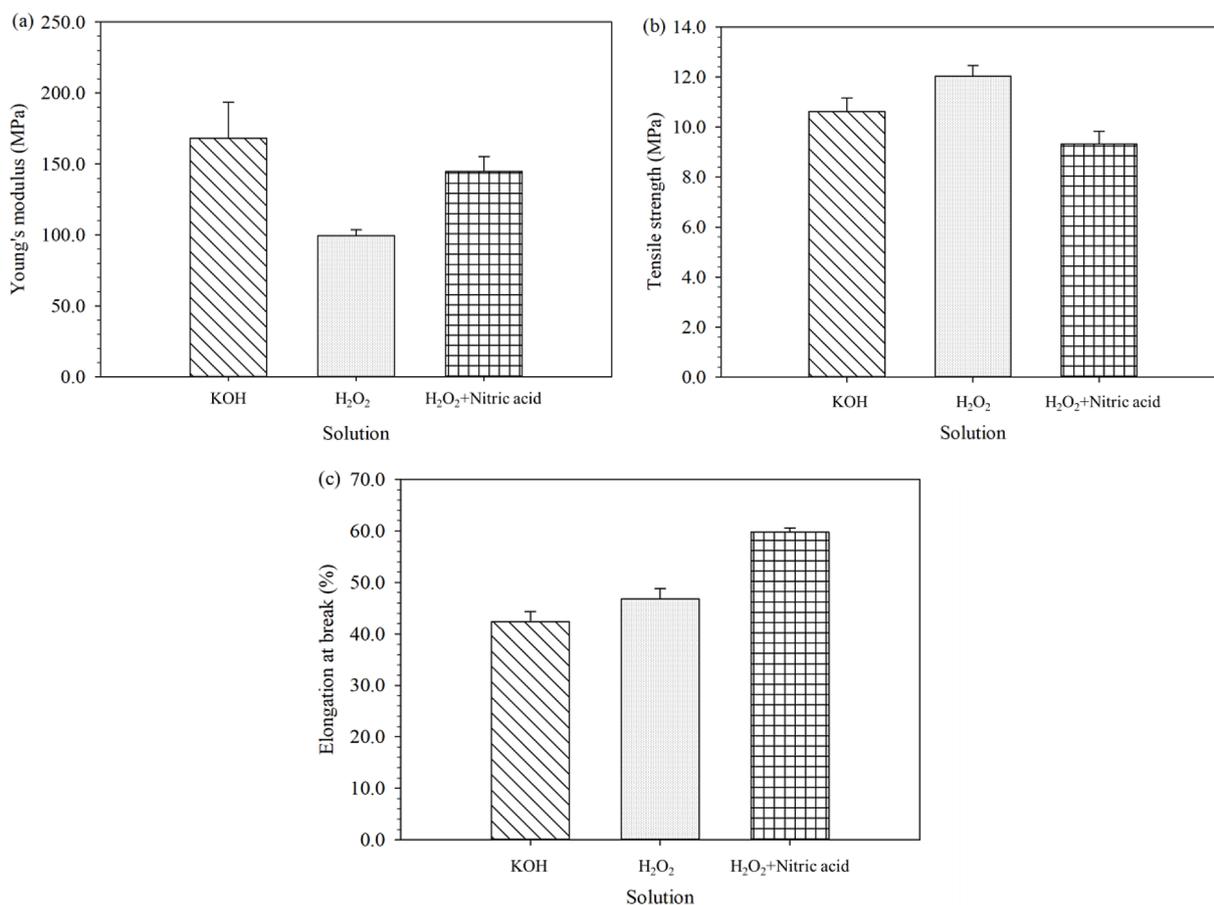


Figure 2. Mechanical properties of PVA/BC films in different solutions for BC preparation

Table 2. The ratio of PVA/BC solution for film preparation

Ratio (PVA:BC)	PVA solution/100 mL		BC solution/100 mL		
	PVA (g)	Glycerol (g)	BC (g)	% Nitric acid	% H ₂ O ₂
100:0	10.0	1.5	0.0	1.0	2.0
90:10	9.0	1.5	1.0	1.0	2.0
80:20	8.0	1.5	2.0	1.0	2.0
70:30	7.0	1.5	3.0	1.0	2.0
60:40	6.0	1.5	4.0	1.0	2.0

strength of the film was 12.04 ± 0.43 MPa. Preparing a PVA/BC film in a solution of 1% nitric acid and 2% H₂O₂ resulted in the optimal elongation at break of $59.78 \pm 0.77\%$. Elongation at break measures the maximum distance the material can stretch to the point where the test piece breaks from each other compared to the initial distance (gauge length). However, the tensile strength value for the 1% nitric acid and 2% H₂O₂ is lower than in other solutions. From the experimental results, the preparation of BC in 1% nitric acid and 2% H₂O₂ solution was selected because it gave high elongation at break, was suitable for making films, and could be used to prepare BC for mixing with PVA well (Ma et al., 2016). The proportion of PVA and BC was changed; the amount of BC was increased to 10, 20, 30, and 40%, as shown in Table 2.

The characterization of PVA/BC films

Structure analysis

The thickness of PVA/BC films obtained from the solution casting method varies from 0.11 to 0.21 mm. as shown in Table 3. The addition of BC impacted film thickness. In research by Xu et al. (2020), adding hemicelluloses and cellulose nanocrystals to a BC also increased thickness compared with a pure PVA film. However, the thickness increase could be a result of increasing the solid content in the film (Pereda et al., 2014; Reddy and Rhim, 2014).

Table 3. The thickness of PVA/BC films

Ratio (PVA:BC)	Thickness (mm)
100:0	0.11±0.00
90:10	0.21±0.00
80:20	0.21±0.01
70:30	0.19±0.01
60:40	0.18±0.00

The surface structure of PVA/BC film was observed by electron microscope scanning, and the different BC rates were compared. It was found that the surface of the film was smooth when adding low BC, as shown in Figure 3c, but it is different from pure PVA (Figure 3b) because PVA and BC still dissolve well and can disperse homogeneously (Choo et al., 2016), thus the film has a smooth surface. With an increase in BC, the roughness of the surface also increased, as shown in Figures 3d, e, and f. The higher the volume of BC, the lower the dispersibility in the film and the solubility effect of BC, resulting in a rough surface. The film has a dehydration process and a strong crystallinity, and phase separation may occur between PVA and BC, resulting in an unsmooth surface (Laxmeshwar et al., 2012). However, it shows that BC can interact at the PVA surface, and increasing the amount of BC can cause voids at the film surface, as shown by the arrow in Figure 3f.

The structure was analyzed by FTIR, and the data is shown in Figure 4. The shapes and positions of most peaks are similar to pure PVA film, and the C-H and C-O bonds peak at 2942 cm⁻¹ (Wang et al., 2019), 1654 cm⁻¹ (C=O stretching), 1031 cm⁻¹ (C-OH stretching), 825 cm⁻¹ (C-C stretching), and the peak at 3268 cm⁻¹ is stretching vibration of O-H (Shao et al., 2016). However, the peak is present in more hydroxyl groups to form intra- and intermolecularly on the PVA backbone by hydrogen bonding (Mansur et al., 2004).

Mechanical properties test

Adding BC affects the mechanical properties by increasing Young's modulus and tensile strength and decreasing elongation at break compared with a pure PVA. The value of Young's modulus of film with a PVA:BC ratio of 60:40 is 595.57 ± 45.51 MPa higher than the pure PVA or 100:0, which is 542.57 ± 49.65 MPa. This indicates that BC can improve deformation resistance or film hardness when added at high doses

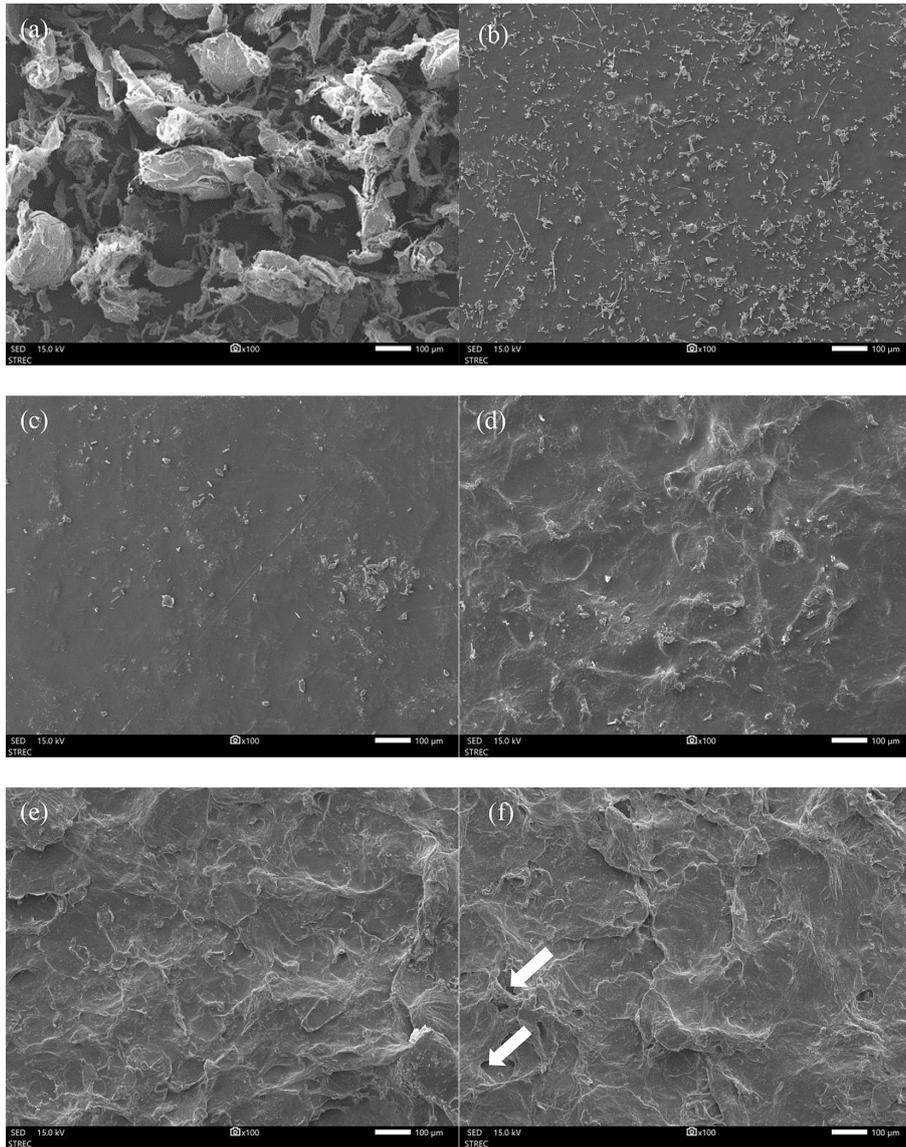


Figure 3. The SEM images of the BC sheet surface (a) and films with different ratios of PVA:BC, specifically 100:0 (b), 90:10 (c), 80:20 (d), 70:30 (e), and 60:40 (f)

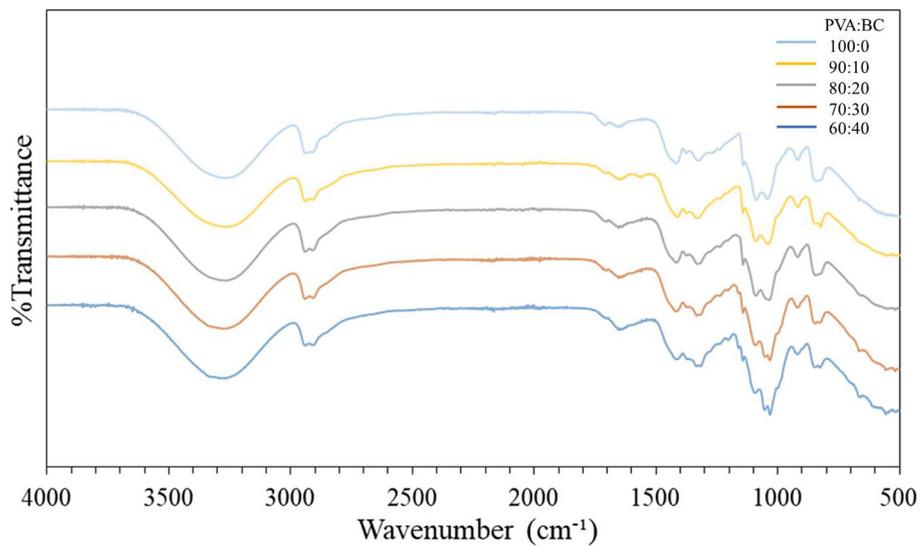


Figure 4. FTIR spectra of PVA/BC films with different ratios between PVA and BC composition

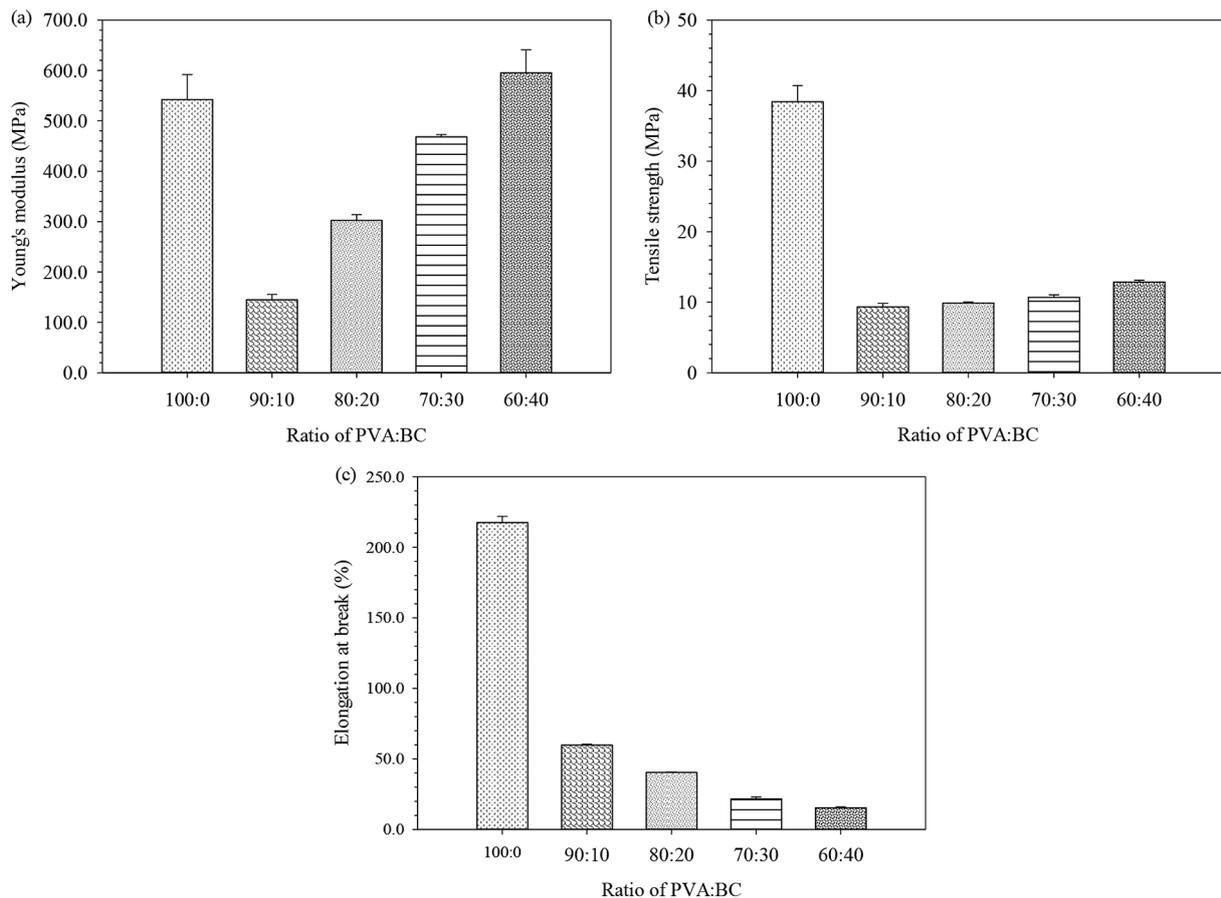


Figure 5. The tensile profiles in terms of Young's modulus (a), tensile strength (b), and elongation at break (c) of pure PVA (100:0) and PVA/BC films reinforced with different ratios between PVA and BC composition (90:10, 80:20, 70:30, and 60:40)

to films. When BC is added at a ratio of 90:10, 80:20, 70:30, and 60:40, the tensile strength increases with 9.32 ± 0.51 , 9.87 ± 0.15 , 10.67 ± 0.36 and 12.83 ± 0.28 MPa, respectively compared to pure PVA. Pure PVA has a much higher tensile strength of 38.42 ± 2.30 MPa. The elongation at break also decreased with increasing BC content. However, the addition of BC, which has a large amount of functional hydroxyl on the structure's surface, interacts with the PVA and glycerol matrix to form hydrogen bonds (Agustin et al., 2013). This affects stiff network structure, strictly limits the chain mobility of the PVA matrix (Mehrali et al., 2013), and results in a tensile strength increase, less flexible or a lower elongation at break value, and lower PVA deformation capacity as compared to pure PVA (Lu et al., 2006; Yang et al., 2010; Choo et al., 2013). In general, if the tensile resistance increases so does the elasticity. Young's modulus indicates that adding BC can be used as a reinforcement material. Results of mechanical properties of films with different PVA:BC ratios are presented in Figure 5.

Water absorption and moisture content

The molecular structure of PVA contains a large number of hydroxyl OH groups, which have hydrophilic properties. It can absorb water within its structure well, but with the addition of BC, PVA absorbed less water (Haghighi et al., 2021). Because the BC is highly crystalline and insoluble when mixed into the structure, it reduces the penetration of water molecules into the film. It also reduces the formation of hydrogen bonds in the OH groups, thereby reducing the film's moisture content. The increase in BC content in the membrane leads to a decrease in water absorption, as shown in Figure 6. The moisture content in the film structure also decreases, starting with the pure PVA film (100:0), which has a water content of $22.48 \pm 0.43\%$. As the BC content increased from 10, 20, 30, and 40%, the moisture percentage decreased to $19.56 \pm 0.21\%$, $18.88 \pm 0.21\%$, $17.69 \pm 0.28\%$, and $15.02 \pm 0.30\%$, as presented in Figure 7. However, the decreased water absorption effect of PVA/BC is maintained when exposed to moisture.

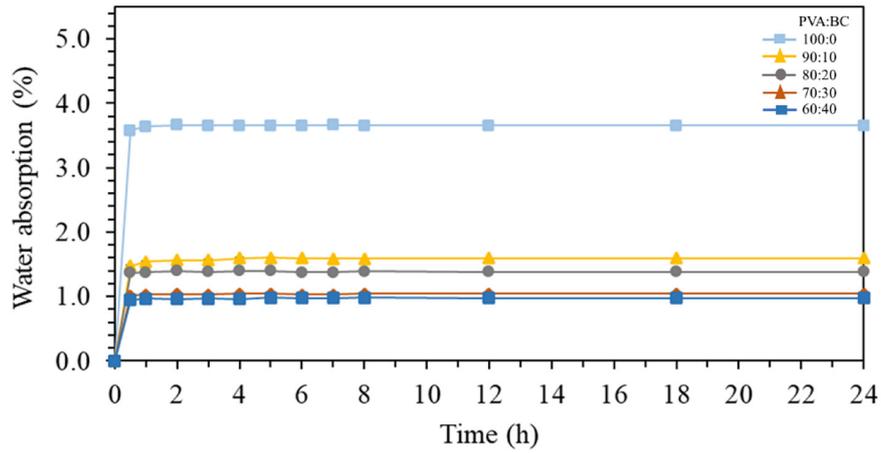


Figure 6. Water absorption of PVA/BC films with different between PVA and BC composition

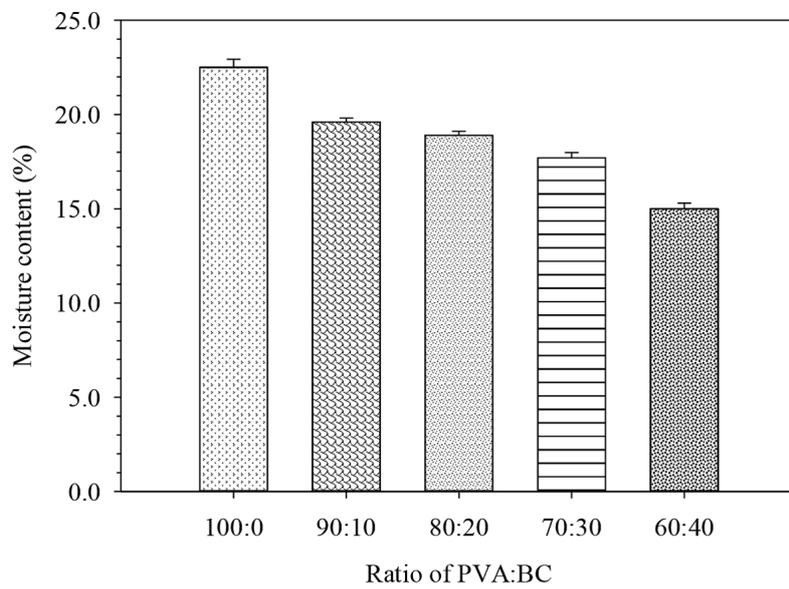


Figure 7. Moisture content of PVA/BC films with different ratios between PVA and BC composition

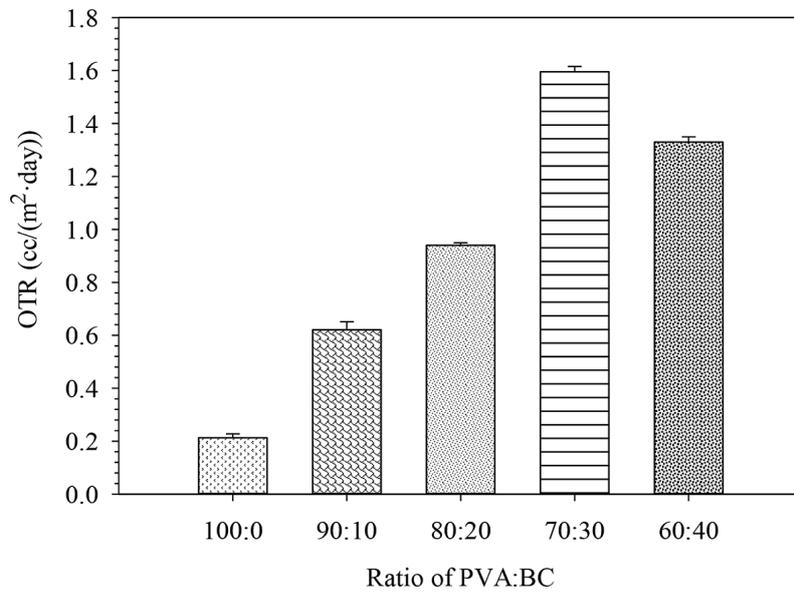


Figure 8. OTR of PVA/BC films with different ratios between PVA and BC composition

Oxygen transmission rates (OTR) and water vapor transmission rates (WVRT)

The OTR was best in the PVA:BC ratio of 70:30, 1.60 ± 0.02 cc/(m²·day). Values of OTR for all ratios are illustrated in Figure 8. For the value of WVTR, all ratios were higher than 1,000 g/(m²·day) because the PVA/BC film had poor water resistance due to the main components in the structure belonging to hydrophilic materials.

Thermogravimetric analysis

PVA/BC film had a slight weight loss in the early stage of thermal analysis, as shown in Figure 9. In the first stage, weight loss occurred gradually from 50–200 °C, possibly due to the loss of moisture and the trapped and adsorbed water in the PVA/BC film (Demchenko et al., 2005). In the second stage, the temperature increased from 200–330 °C, and the BC in the film decomposed

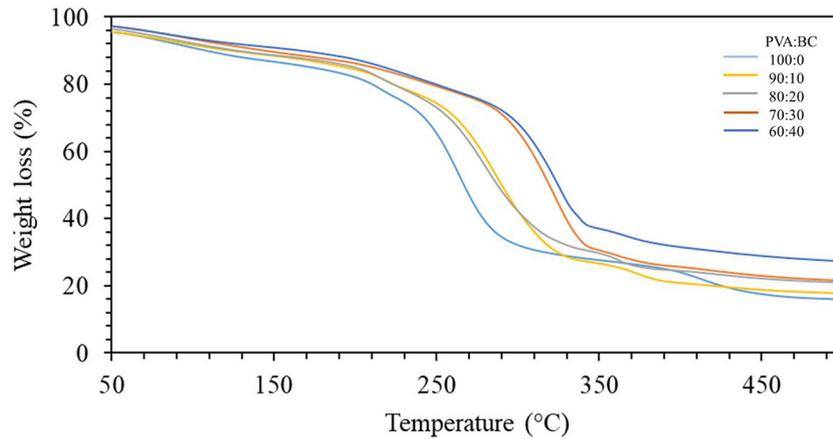


Figure 9. Thermal stability analysis curves of PVA/BC films with different ratios between PVA and BC composition

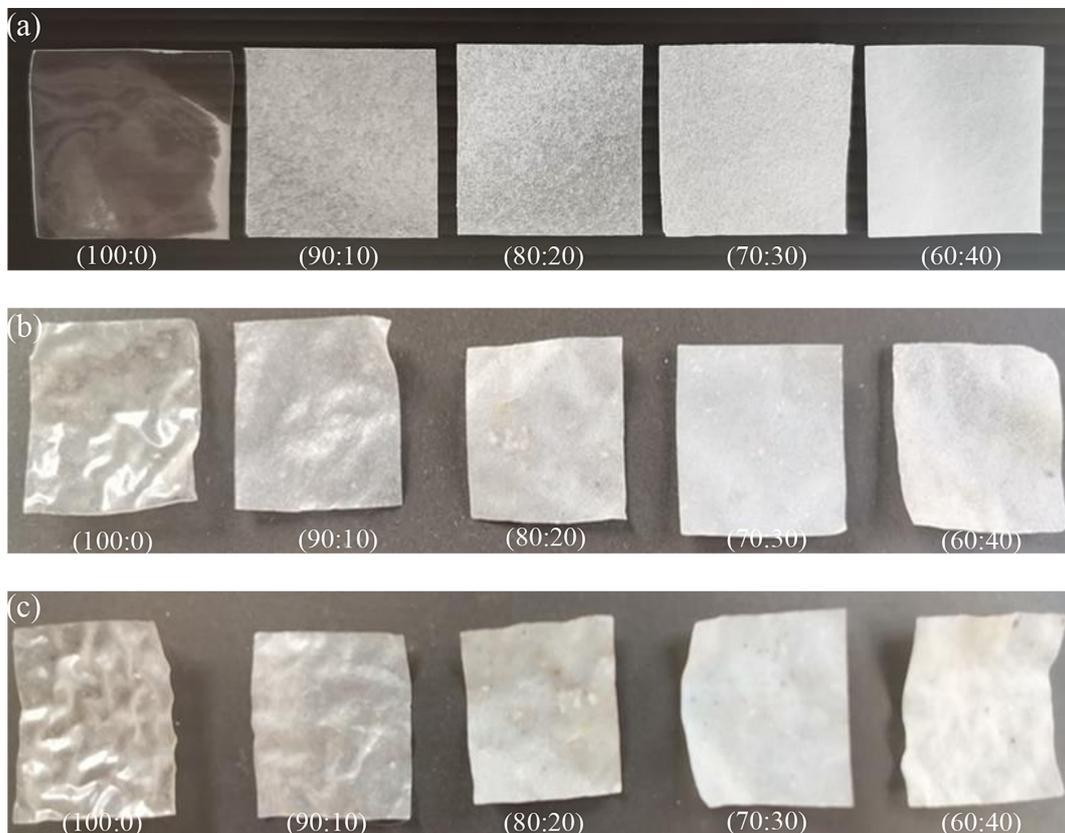


Figure 10. Soil burial biodegradation test of PVA/BC films in different ratios. Conditions shown are before degradation (a) and after 7 days (b) and 14 days (c)

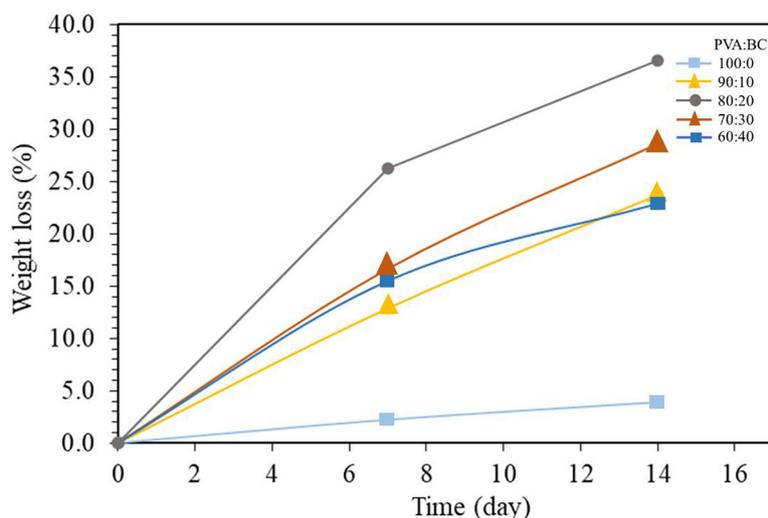


Figure 11. Biodegradation rate of PVA/BC films in different ratios

rapidly. Weight was reduced by approximately 30% through pyrolysis (Nigam et al., 2021). In the final stage, the temperature was at 330 °C. The decrease in weight shows that the thermal degradation of the PVA/BC is lower than pure PVA film. TGA data showed that the content of BC increased with decreasing weight loss.

Biodegradation test

The biodegradability of PVA/BC film is shown in Figure 10 and was assessed based on weight loss. The initial weight loss was 0%. After 7 and 14 days of testing, it was found that every PVA/BC film treatment degraded, as shown in Figure 11. The decomposition rate increased over time, considering the increase in percentage weight loss. Increasing the BC content improved film degradation compared to pure PVA film (100:0), with only $3.94 \pm 1.42\%$ degradation after 14 days. The best degradation rate over 14 days was 80:20, with a weight loss of $36.57 \pm 1.26\%$.

The degradation rate increased because soil microorganisms involved in degradation are adapted to pre-developed conditions. (Meit et al., 2020). When microorganisms in the soil are adapted to the environment, they grow and digest film rapidly. The results showed that the PVA/BC film was biodegraded spontaneously, and a higher BC content also helped improve the biodegradability of the film.

CONCLUSIONS

Structural characteristics – the addition of BC made the film thicker and increased the

surface roughness of the film. Mechanical properties – the study found that when adding BC, the film had a lower elongation at break value than pure PVA. As the BC increased, the elongation at break decreased. The tensile strength decreases with the addition of the BC, related to Young's modulus, and is decreased after adding the BC. However, when adding BC at a high ratio of 60:40, it had values similar to pure PVA. Therefore, from the high Young's modulus, BC can be used as a reinforcement in PVA to make the film have good stability properties. Water absorption and moisture content – the addition of BC made the film absorb less water, resulting in a lower moisture content. With an increase in BC, water absorption and moisture content further decreased. Therefore, the addition of BC made the film insensitive to moisture.

Oxygen transmission rates – the OTR was very high in the case of BC addition compared to pure PVA, but when BC is added, OTR decreases by up to 40%. This is high in terms of WVTR.

Thermal stability by thermogravimetric analysis – results showed that adding BC reduced the film's degradation or stabilized it at high temperatures. Biodegradation properties – the addition of BC helps to improve the biodegradability of the film. The higher the amount added, the higher the biodegradation rate. Moreover, PVA/BC was successfully prepared with a simple solution casting method. PVA blended with BC can improve its biodegradability properties. Also, BC material can help reduce composite material costs. PVA/BC is thus another environmentally friendly material.

Acknowledgments

This study was supported by a scholarship from the Thailand Science Research and Innovation (TSRI) to Srinakharinwirot University (Code; 004/2565). We would like to thank the Agricultural Product Innovation and Technology for supporting the analysis instruments.

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