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Poly(butylene adipate-*co*-terephthalate)/thermoplastic canna starch (*Canna edulis ker.*) (PBAT/TPS) blend film – A novel biodegradable material

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ABSTRACT

The PBAT (poly (butylene adipate-*co*-terephthalate) is a promising biodegradable material. However, it is often blend with hydrophilic polymers since its degradation rate in the aquatic environment is still limited. In this study, the blend PBAT/TPS (thermoplastic starch) films, namely BFs, were prepared by a blow extrusion approach, and evaluated for hydrolysis in four studied mediums acid (HCl, 1 M, 2 M, and 3 M), alkaline (NaOH, pH = 9, 11, and 13), phosphate buffer (pH = 7.4), and artificial seawater. The hydrolyzed BFs were characterized by weight loss, mechanical properties, scanning electron microscopy (SEM), Fourier transform infrared spectra (FTIR), and differential scanning calorimetry (DSC). A larger starch content in the BFs caused hydrolysis more quickly. The highest hydrolytic rate was found in the alkaline solution, followed by the acid medium. The complete abiotic hydrolysis of the BFs was 3 M HCl for 14 days or NaOH (pH 13) for 35 days. After 180 days of incubation, the film containing 70.5 % PBAT/TPS granules has been associated with the highest biodegradation rate of 76.31 % in composting.

1. Introduction

Plastic pollution has risen globally as a result of the widespread use of plastics in many different applications, such as packaging and mulching films [1]. As can be seen, biodegradable materials were widely applied in various industrial sectors, which usefully replaced traditional plastics, and significantly reduced the quantity of white pollution [1,2]. Currently, various biodegradable polymers have successfully been created, such as poly (lactic acid), poly (butylene succinate-*co*-butylene adipate), polyhydroxyalkanoates, poly (ε-caprolactone), and poly (butylene succinate), especially poly (butylene adipate-*co*-terephthalate) (PBAT) [3–8]. The PBAT is one of the most widely used biodegradable polymers for environmentally friendly biodegradable products, such as food packaging, shopping bags, agricultural mulch films, and garbage bags [9]. However, because of the relatively high price, the PBAT needs to be combined with other polymers to reduce production costs [10]. Synthesis of the PBAT/starch blends has drawn much interest recently since they are considered as an alternative for petroleum-derived polymers, which directly contacted with food. In addition, the studies on the PBAT/starch blends were mainly concentrated on popular starches, such as cassava, corn, potato, and canna [11–14]. For instance, by a blend film between the PBAT and cassava starch, the light transmittance and mechanical properties of its membrane decreased [11].

Canna (*Canna edulis* Ker.) is cheap, and has a high starch content (>95 %). In Vietnam, canna has been growing in numerous regions with over 30,000 ha, and annual production of about 300,000 tons. Canna starch-based films have good mechanical and biodegradable properties, and a low vapor permeability. Thereby, it brings out the potential in the use of starch for biodegradable films [15]. To date, there has been no study on the biodegradable films based on canna starch.

The biodegradable polymers can be the opposite materials of the typical non-biodegradable polymers, which were possibly composted [16]. The degradation of the biodegradable polymers has included two primary degradable steps, and one ultimate mineralization [17–19]. Primary degradation is also known as abiotic hydrolysis. Water appeared in the system for the hydrolysis reaction. There are three different kinds of hydrolysis, including the base-catalyzed hydrolysis,

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the acid-catalyzed hydrolysis, and the autocatalytic hydrolysis [17]. The principle of all three kinds is that the water is used to break the ester bonds, thereby resulting in reduced molecular weight fragments [18]. Ultimate mineralization by microorganisms also contained three main steps: (i) colonization of the polymer surface by microorganisms, (ii) secretion of extracellular microbial enzymes that depolymerize the polymer into low-molecular-weight compounds, and (iii) microbial uptake and utilization of these compounds, incorporating polymer carbon into biomass or releasing it as CO_2, etc [19]. The ultimate aims of this study are to evaluate the abiotic hydrolysis and biodegradation of the blend PBAT/TPS films (BFs) in different conditions.

2. Experimental

2.1. Materials

The PBAT resin was supplied by SMBEST Pvt. Ltd. (Korea) with a density of 1.22 g/cm^3, and a melt flow index (MFI) of 3–5 g/10 min at 2.16 kg/190 °C. Canna starch (*Canna edulis* Ker.) was isolated from fresh canna, according to a previous report [20]. The obtained canna starch contained 33 % amylose, 60 % amylopectin, 0.22 % ash, and 0.31 % crude fiber, without lipid and protein. Epoxidized soybean oil was supplied by Thang Long Chemical., JSC (Vietnam) with a density of 0.98 g/cm^3, and an oxirane index of 6.5 %. Polycaprolactone (PCL) with a density of 1.14 g/cm^3 and a melting temperature (T_m) of 60–62 °C was used as a processing aid, while Erucamide with the T_m of 80–85 °C was used as a slip agent. All chemical reagents were purchased from Sigma.

2.2. Preparation of thermoplastic canna starch

Canna starch (dried at 70 °C for 12 h) and glycerol (70/30, w/w) were mixed on a high-speed mixer, and then fed into the TSE-52 co-rotating twin-screw extruder with the temperature zone of 110–160 °C and the speed of 70 rpm to obtain the TPS.

2.3. Preparation of the BFs

To improve the dispersion of the TPS in the PBAT matrix during the film-blowing process, the first PBAT/TPS (PBAT, TPS, and ESO, 40/60/5, w/w/w) blend granules were prepared using a twin-screw extruder (model SJSL-36/600–18.5-48) with an L/D ratio of 48:1, w/w, the temperature zone of 110–190 °C and the speed of 90 rpm.

The blend BF film with a thickness of 25 μm was obtained by blown extrusion using a twin-screw extruder (model GBCE-50/650) with heating zones of 155–180 °C. The labels for the samples were summarized in Table 1.

2.4. Abiotic and biotic degradation experiments

2.4.1. Abiotic hydrolysis tests

Samples were hydrolyzed in the following mediums.

- HCl solution with different concentrations (1 M, 2 M, and 3 M).
- NaOH solution with different pH (9, 11, and 13).
- Phosphate buffer solution with pH = 7.4 (mixture KH_2PO_4 0.1 N + NaOH 0.1 N).
- Artificial seawater (1 L of artificial seawater consists of 35 g NaCl, 1.153 g $CaCl_2$, 0.72 g KCl, 0.198 g $NaHCO_3$, 0.0958 g H_3BO_3, 2.26 g $MgCl_2$, 3.248 g $MgSO_4$, 0.0624 g $NaSiO_3$, and 0.013 g $AlCl_3$).

Table 1
The biodegradable BFs formulations.

Sample	Composition (%)			
	PBAT granules	PBAT/TPS granules	PCL	Erucamide
BF0	94	0	5	1
BF20	47	47	5	1
BF30	23.5	70.5	5	1
BF40	0	94	5	1

The film samples were cut into 3 × 13 cm pieces, were hydrolyzed in 200 mL of four different solutions at room temperature, protected from sunlight, and in a stationary state. All samples were stored in sealed containers to prevent interaction with the environment. For each period of three days, the samples were washed with distilled water, then were dried and evaluated for weight loss, morphology, structure, and mechanical and thermal properties.

Weight loss: The weight loss of the films (w%) during hydrolysis was calculated using equation (1):

$$w(\%) = \frac{m_0 - m_s}{m_0} \times 100 \quad (1)$$

Where m_o is the initial mass of the sample, and m_s is the mass of the sample after hydrolysis.

Morphology: A scanning electron microscope (SEM) (JEOL 6490, Japan) was used to view the surface morphology before and after hydrolysis. The surface of the films was coated with a thin layer of platinum before measuring.

Mechanical properties: By the standard ASTM D882, tensile properties were determined using the BP-1068 Testing Machine (China) at a crosshead speed of 10 mm/min [21]. All the tested samples were conditioned at 25 °C for 24 h before testing. An average value of five tests was reported.

Fourier transform infrared spectroscopy (FTIR): The chemical structure of BFs was studied by using the FTIR spectrometer (Nicolet iS10) with a resolution of 4 cm^{-1}, and scanning range from 4000 to 500 cm^{-1}.

Differential scanning calorimetry (DSC): The DSC (NETZSCH DSC 204F1 Phoenix, Germany) was used to determine the thermal properties of the film samples before and after hydrolysis. The samples were heated from room temperature to 200 °C at 10 °C/min under N_2 atmosphere. The relative crystallinity (X_c) was calculated using equation (2):

$$X_c = \Delta H_m \text{ x } 100\%/(f \text{ x } \Delta H_m^o) \quad (2)$$

Where ΔH_m is the melting enthalpy in the experiment, f is the weight fraction of the PBAT in the blend films, ΔH_m^o is the perfect enthalpy of 100 % crystalline PBAT (=114 J/g) [22].

2.4.2. Biodegradation experiments

The aerobic biodegradability of the BFs under the composting conditions was determined by measuring the amount of CO_2 emitted when the samples were degraded, according to the international standard ISO 14855–1:2012 (Fig. 1) [23]. The degradation reactor was placed in a thermostatic water tank of 58 ± 2 °C. A CO_2-captured trap (saturated solution of NaOH). The amount of CO_2 was determined by titration with a 0.1 M HCl standard solution for three days. Experiments were performed in triplicate for 180 days. Cellulose was used as a reference. The blank reactor without sample was carried out similar.

The biodegradable percentage (D_t) of each test material was calculated according to equation (3):

$$D_t = \frac{(CO_2)_T - (CO_2)_B}{ThCO_2} \times 100 \quad (3)$$

Where $(CO_2)_T$ is the cumulative mass of CO_2 evolved by the flask containing sample (g), $(CO_2)_B$ is the cumulative mass of CO_2 evolved by the blank flask (g), $ThCO_2$ is the theoretical mass of CO_2 produced by the test material (g).

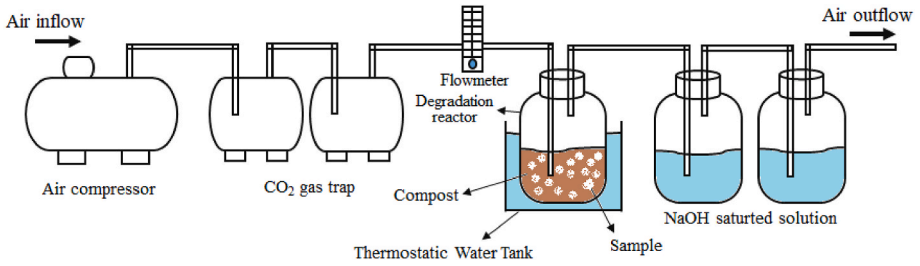

Fig. 1. Schematic diagram of the biodegradability test.

3. Results and discussion

3.1. Hydrolysis of the BFs

3.1.1. Weight loss

The weight loss of BFs in the studied media is shown in Figs 2-4. Regarding the HCl solution, two processes could take place: Starch hydrolysis and PBAT hydrolysis. The higher starch content in films and the higher HCl concentration lead to the greater weight loss of the films. The film without starch (BF0) degraded the slowest, the weight loss of BF0 was only 26.85 % after 49 days in 1 M HCl solution, 36.58 % after 42 days in 2 M HCl solution, and 33.79 % after 35 days in 3 M HCl solution. The weight loss of the films with starch was 39.64–55.26 % after 49 days in 1 M HCl solution, 48.55–69.74 % after 42 days in 2 M HCl solution, and 52.79–72.15 % after 35 days in 3 M HCl solution. Apparently, starch was swollen up easily, and hydrolyzed in an acidic solution. Therefore, micro-holes appear, which allowed water and H^+ ions to diffuse more easily, thereby the PBAT was easily hydrolyzed. The higher the H^+ concentration was, the stronger this process happened [24]. Raghava et al. (2001) also obtained similar results when studying the hydrolysis of PLA/PE/starch composites in HCl solution [25].

The mechanism is associated with the protonation of ester groups by electrophile agents, such as H_3O^+, resulting in the cleavage of the acyl-oxygen bond, and the products were oligomers of adipic acid (AA), terephthalic acid (PTA) or the PBAT fragments containing terminal –COOH groups (Fig. 5a. These products then acted as "self-catalysts" for hydrolysis of the PBAT in the acidic solution. Thus, the lower the HCl concentration was, the less H^+ was produced to protonate the ester group, leading to a decrease in the number of nucleophile centers and slower reactions [26]. After 35 days of hydrolysis in 3 M HCl solution, the blend films were degraded into small pieces that could not be recovered. Therefore, the BFs after 35 days at 3 M HCl concentration were used to evaluate the changes in morphology and structure.

Similar to the acidic solution, the weight loss of BFs in the alkaline medium was greater with longer time treatment, and higher starch content in the film (BF40 > BF30 > BF20 > BF0). The weight loss of the BFs was 69.47–88.58 % after 18 days in NaOH solution (pH 13),

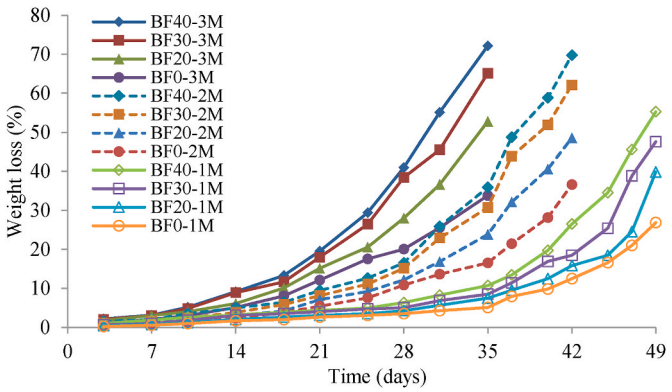

Fig. 2. The weight loss of BFs in HCl solution.

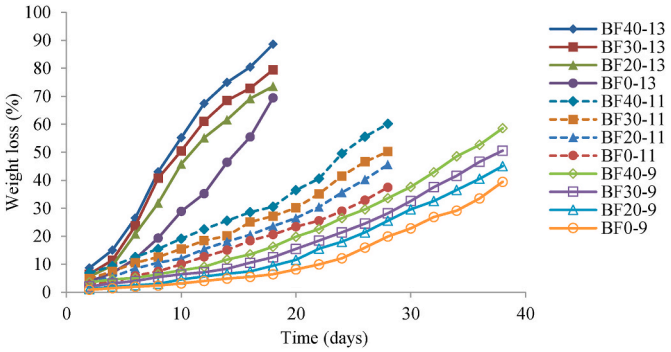

Fig. 3. The weight loss of BFs in NaOH solution.

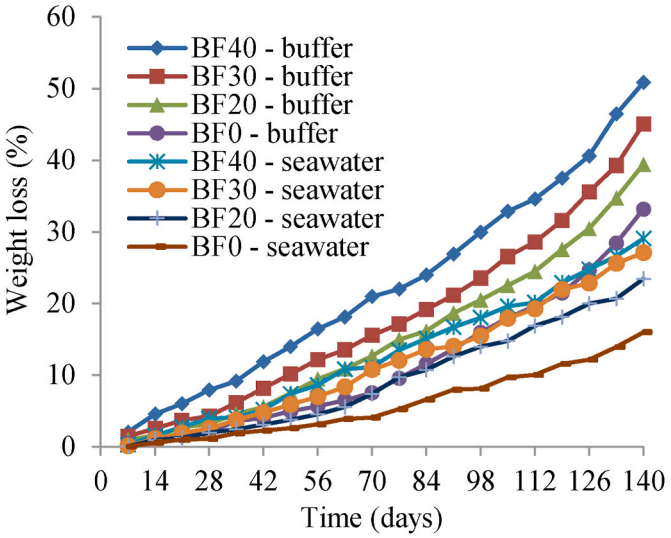

Fig. 4. The weight loss of BFs in phosphate buffer solution and artificial seawater.

37.45–60.14 % after 28 days in NaOH solution (pH 11) and 38.41–58.56 % after 38 days in NaOH solution (pH 11). The BFs were degraded promptly by increasing the pH of the NaOH solution, in which starch was quickly hydrolyzed and dissolved [27]. In this case, OH groups in the alkaline medium acted as a strong nucleophile agent to attack C=O groups (Fig. 5b) [26]. The rapid dissolution of the sodium salts (which was formed by hydrolysis, created surface defects) that allows the water molecules and OH^- ions to diffuse more easily, thereby the PBAT is more hydrolyzed. After 14 days of hydrolysis in the NaOH solution (pH 13), the blend films were degraded into small pieces that could not be recovered. Therefore, the BFs after 14 days at pH 13 were used to evaluate the morphological and structural changes.

From Fig. 4, the BFs were hydrolyzed faster in buffer solution than in artificial seawater. After 140 days, the weight loss of BFs was

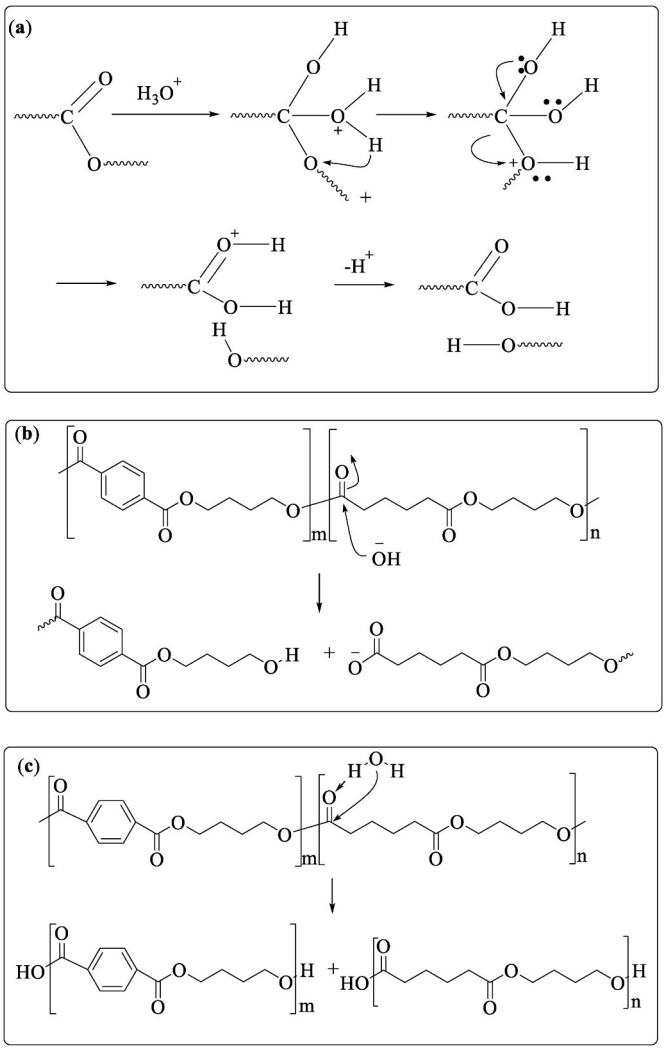

Fig. 5. Hydrolysis mechanism of PBAT in acid (**a**), alkaline (**a**), and neutral media (**c**).

33.15–50.82 %, and 15.97–29.05 % in phosphate buffer solution, and artificial seawater, respectively. The 140-day treatment could be seen as an optimal condition since the weight loss was observed insignificant after this period. As can be seen that the weight loss of the films also increased with their starch content. Water was easily absorbed by starch [28], Hence, the more starch amount the film loaded, the more water would be absorbed. This promoted the diffusion of water molecules inside the material [29], and created favorable conditions for the hydrolysis of the BFs due to the higher hydrophilicity of the films. Stoukal et al. (2015) suggested that the abiotic hydrolysis could be accelerated by increasing the hydrophilicity of the material's surface, which facilitated the penetration of water molecules into the material in the early stages of hydrolysis [18].

The hydrolysis rate of the BFs in buffer solution and artificial seawater was much slower than in the acidic and alkaline solutions. This is because the PBAT hydrolysis was a self-hydrolysis owing to the terminal COOH group acting as a catalyst, hence the PBAT hydrolysis was slow. Water molecules were also relatively weak nucleophile agents in the neutral mediums, thereby being difficult to attack C═O groups [26]. The results also showed that the BFs were hydrolyzed in alkaline solution faster than the remaining solutions. This was because the hydrolysis of PBAT was an irreversible reaction in alkaline solutions, while it was a reversible reaction in acidic and neutral solutions.

3.1.2. Surface morphology

The surface morphology of the films before and after hydrolysis in 3 M HCl, NaOH pH 13, buffer solution, and artificial seawater was shown in Figs 6-9, respectively. The SEM images of the original BFs showed that the starch particles were dispersed into the PBAT matrix quite uniformly, the TPS and PBAT matrix did not exhibit any phase separation, indicating a good compatibility between the two phases. The surface of the starch-free PBAT film (BF0) was relatively smooth.

The changes in the surface of the hydrolyzed BFs were in a visible order of alkaline > acid > phosphate buffer > artificial seawater. The BFs' surface appeared voids and pores. The number and size of pores increased in proportion to how much starch the films loaded. According to Shujun et al. (2005), the main component of the blend formed the continuous phase, while the lesser component was the dispersed phase [30]. In this case, the continuous phase was formed by the PBAT polymer and the TPS was the dispersed phase. The starch molecules in the blend film, which were hydrolyzed and dissolved in aqueous solution, might be responsible for the porosity that developed during hydrolysis [31]. These pores created conditions for water molecules and H^+ ions to diffuse deep into the material, promoting the hydrolysis of the film to take place faster.

In alkaline solution, it is also observed that the surfaces of films were corroded deeper and more widely. This was because, in the alkaline solution, films based on PBAT were corroded by both mechanisms: surface and bulk corrosions, but in the acidic and neutral solutions, only bulk corrosion occurred. Cong et al. (2010) also obtained similar results in a study of hydrolysis of PLA/EVA/$CaCO_3$ composite in HCl, NaOH, and phosphate buffer solutions [26].

3.1.3. Mechanical properties

The changes in tensile strength and elongation at break of the BFs upon hydrolysis in acid, alkaline, phosphate buffer, and artificial seawater were shown in Fig.s 10–12, respectively. The results showed that the tensile strength and elongation at a break of the samples decreased with time. After hydrolysis in 3 M HCl solution, the mechanical properties of films decreased rapidly, followed by 2 M HCl solution, and then slowly by 1 M HCl solution (Fig. 10). It was probable that the amount of H^+ produced by the acid was insufficient to protonate the ester linkage at the low concentrations [26]. The hydrolysis of starch, on the other hand, was also directly influenced by the acid concentration; the greater the acid concentration was, the easier it was for starch to hydrolyze and separate from the PBAT resin matrix, leading to defects on the film surface (as seen in the SEM images). As a result, the mechanical properties decreased rapidly. The results also showed that the higher the starch content in the film was, the faster the mechanical properties decreased, which was consistent with the weight loss. The mechanical properties of the films were reduced during hydrolysis because the defects on the film were produced in proportion to the increased starch content in the film [27].

In the same manner, the mechanical properties of the films decreased with increasing starch content and concentration of NaOH solution (Fig. 11). However, the mechanical properties in NaOH solution decreased much faster than in HCl solution. This was also consistent with the SEM images, the BFs' surface changed strongly when hydrolyzed in NaOH solution.

Similar to the acidic and alkaline mediums, the mechanical properties of the films gradually decreased with longer hydrolysis time in phosphate buffer and artificial seawater (Fig. 12). However, the mechanical properties of films decreased much more slowly than in acidic and alkaline solutions. The higher the starch content of the BFs was, the lower the mechanical properties of the hydrolyzed films were (BF40 < BF30 < BF20 < BF0). The mechanical properties of BF decreased from 31.88 to 60.29 % and from 16.83 to 48.21 % after 140 days of hydrolysis in the mediums phosphate buffer and artificial seawater, respectively. While these values decreased from 57.02 to 89.99 % after 16 days of hydrolysis in pH 11 NaOH solution and 44.78–90.11 % after 18 days of

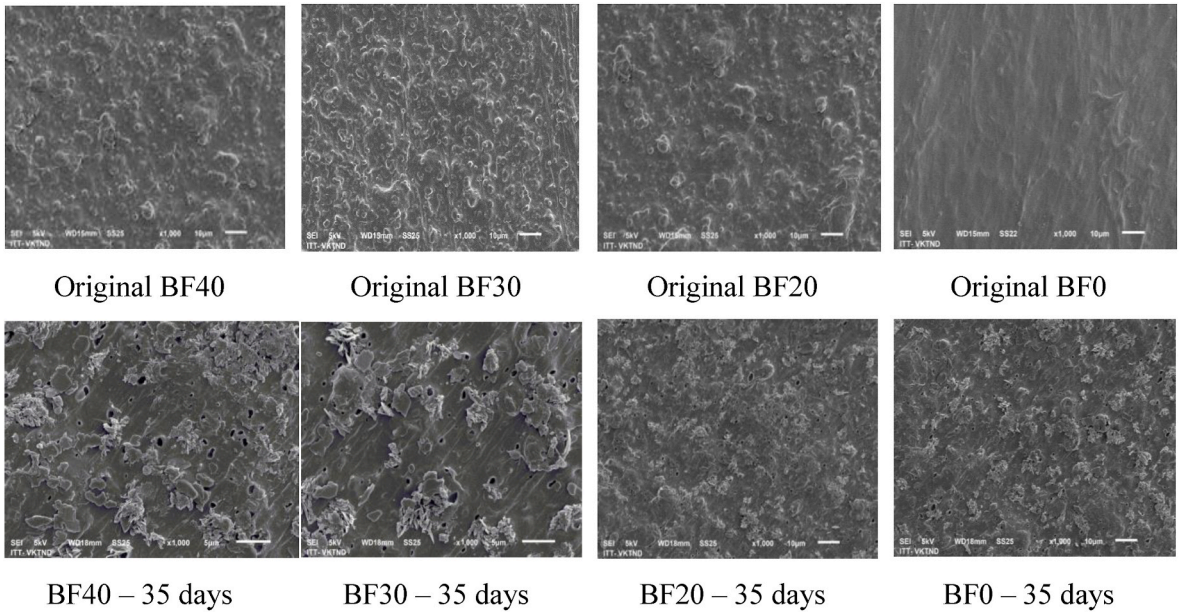

Fig. 6. SEM images of the BFs before and after 35 days of hydrolysis in 3 M HCl solution.

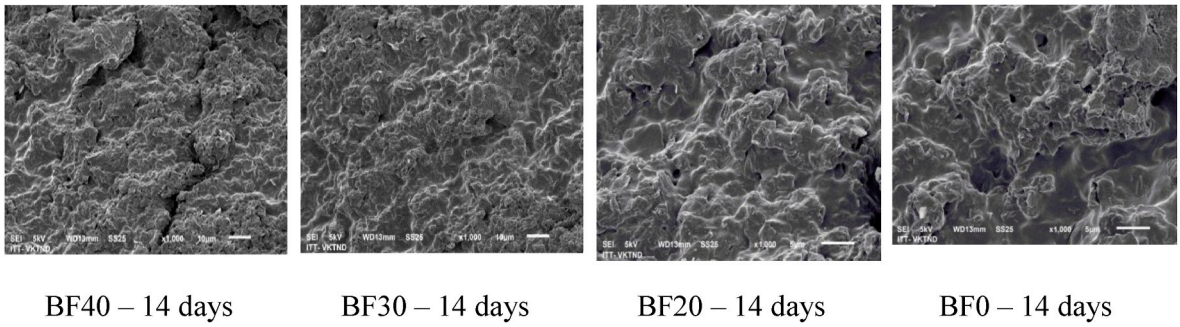

Fig. 7. The SEM images of the BFs after 14 days of hydrolysis in NaOH solution (pH = 13).

hydrolysis in 3 M HCl solution.

3.1.4. The FTIR analysis

Fig. 13 shows the IR spectra of the BF30 films before and after hydrolysis in 3 M HCl solution (35 days), pH 13 NaOH solution (14 days), and phosphate buffer solution and artificial seawater (140 days). A board peak at 3383 cm^{-1}, which was assigned to the stretching vibration of the O–H group in starch (free OH group, intramolecular and intermolecular O–H bonds), could be observed in the IR spectrum of the original BF30 film. The peak at 2924 cm^{-1} was assigned to the stretching vibration of the CH groups in the starch and PBAT molecules [32,33], and the peaks at 1716 and 1273 cm^{-1} were assigned to the stretching vibrations of C═O and C–O groups in aromatic esters, respectively [34,35]. The peaks at 876 and 1018 cm^{-1} were assigned to the bending vibration of the phenyl ring of PBAT and 1105 cm^{-1} corresponded to C═O left-right symmetric stretching vibration absorption [36].

Several functional groups, including OH and C═O of the PBAT structure, can be used as signals to study degradation [37]. The IR spectra of the BF30 film after hydrolysis in HCl, NaOH and phosphate buffer solution did not contain a peak at 3383 cm^{-1}. In artificial seawater, a weak intensity peak at 3383 cm^{-1} in the hydrolyzed films was observed. The characteristic absorption peak of the CH groups was found to range from 2924 to 2925 cm^{-1} with weaker intensities. This demonstrated that very little starch was left in the film structure hydrolyzing in solution. The peaks of the C═O groups was found in the range of 1716-1712 cm^{-1} (in acidic solution), and 1714 cm^{-1} (in alkaline, buffer, and artificial water) with the decreased intensities. Similar results were observed in hydrolysis of the PBAT/TPS, PLA/TPS, and polyvinyl alcohol (PVA)/TPS in a phosphate buffer solution [26]. This is due to the hydrolysis of the PBAT in the blend film, which resulted in the breakdown of the ester linkages and BAT chains, to create shorter PBAT fragments [38]. After hydrolysis, the other peaks also have weaker intensities.

3.1.5. Thermal property

The thermal properties of the BF30 films before and after hydrolysis in four studied mediums were summarized in Table 2. The DSC curves of the films are shown in Fig. 14. It could be seen that the melting temperature (T_m), melting enthalpy (ΔH_m), and crystallinity X_c of the films increased after hydrolysis in an order of seawater < phosphate buffer < acid < alkaline medium. This result can be explained by the environmental effects on the PBAT. In the acidic and alkaline solutions, the PBAT molecules could be cleaved easily into shorter chains, which were easier to arrange in more stable and ordered structures, resulting in the higher crystallinity of the films. In addition, the PBAT is a semi-crystalline polyester, and preferentially decomposed in the amorphous area first [39,40]. The PBAT consists of the rigid butylene terephthalate (BT) units and the soft butylene adipate (BA) units [41]. The amorphous region of PBAT was mainly formed by aliphatic BA units, which was more easily degraded than the major components of the crystalline area (aromatic BT units). Therefore, the decomposition process reduced the amorphous region content and increased the crystalline concentration of

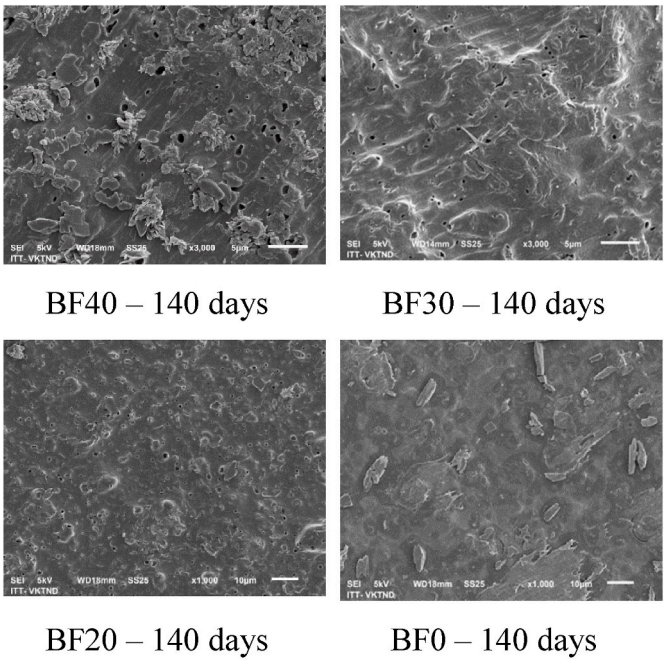

Fig. 8. The SEM images of the BFs after 140 days of hydrolysis in phosphate buffer solution (pH = 7.4).

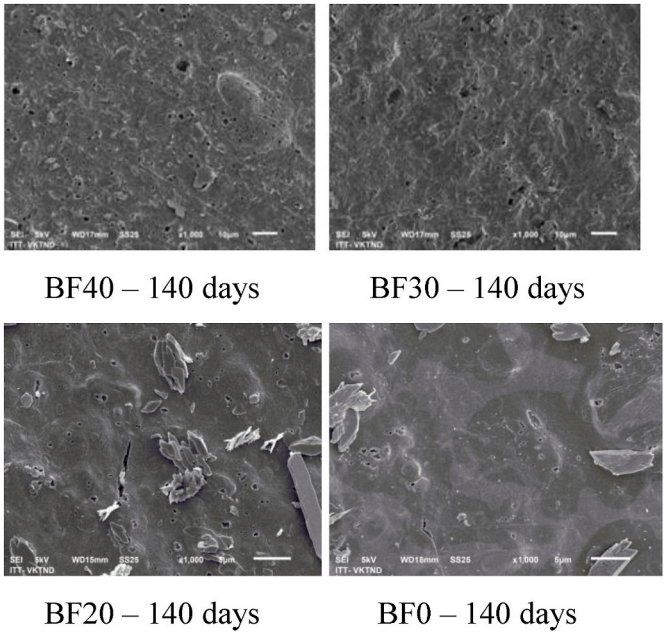

Fig. 9. The SEM images of the BFs after 140 days of hydrolysis in artificial seawater.

the samples. By hydrolysis of the PBAT film in phosphate buffer solution, the amorphous region was first decomposed, resulting in a better crystalline structure with fewer defects [37].

3.2. Biodegradation of the BFs

Evaluating the level of biodegradability is an important step to estimate the final decomposition of materials in the environment. The mineralization rates of BF30, BF20, BF0, and cellulose are shown in Fig. 15. The results showed that the mineralization level of cellulose increased rapidly during the initial incubation, and reached about 84 %

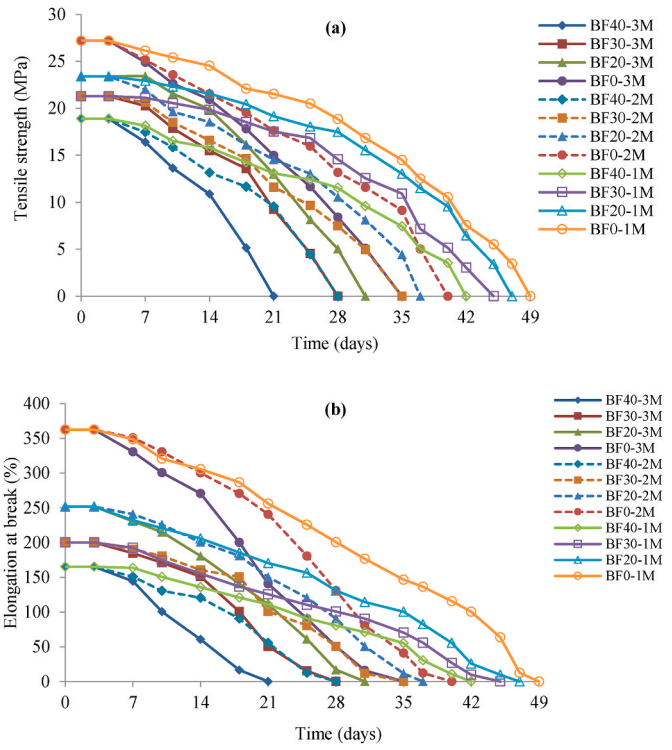

Fig. 10. Tensile strength (a) and elongation at break (b) of the BFs upon hydrolysis in HCl solution.

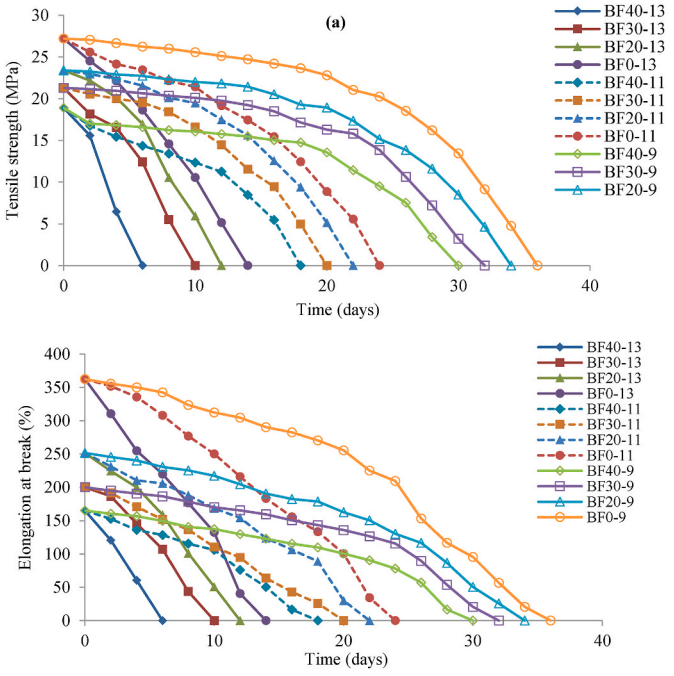

Fig. 11. Tensile strength (a) and elongation at break (b) of the BFs upon hydrolysis in NaOH solution.

after 70 days of incubation. After 180 days, the referenced cellulose reached about 100 % of mineralization. It could be seen that the biodegradation degree of the BFs increased with an increase in the incubation time. In the initial days, the biodegradation degree of the BFs was not significant. Reasonably, the long polymer chains were disrupted shortly at this stage, which facilitated the mineralization [42].

The biodegradation levels of the BF30 and BF20 are faster than that

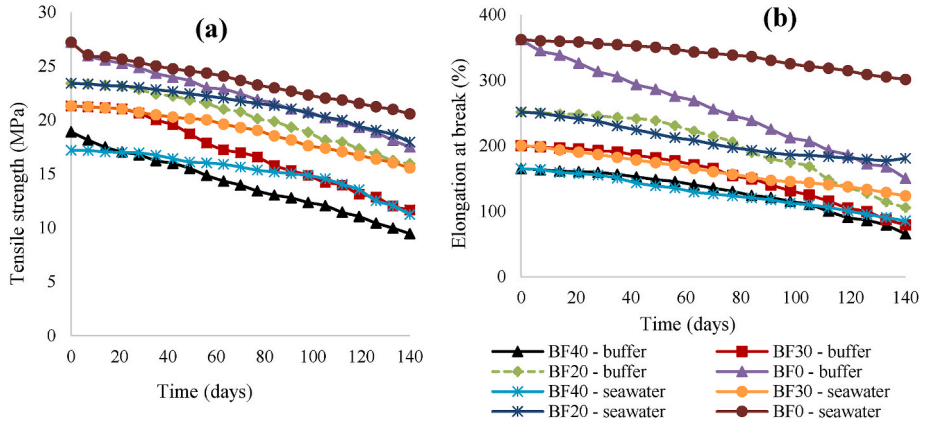

Fig. 12. Tensile strength (a) and elongation at break (b) of the BFs upon hydrolysis in phosphate buffer solution (pH = 7.4) and artificial seawater.

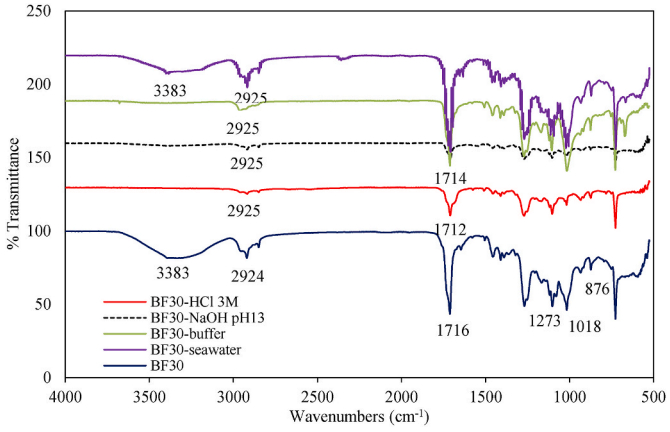

Fig. 13. The IR spectra of the BF30 film before and after hydrolysis.

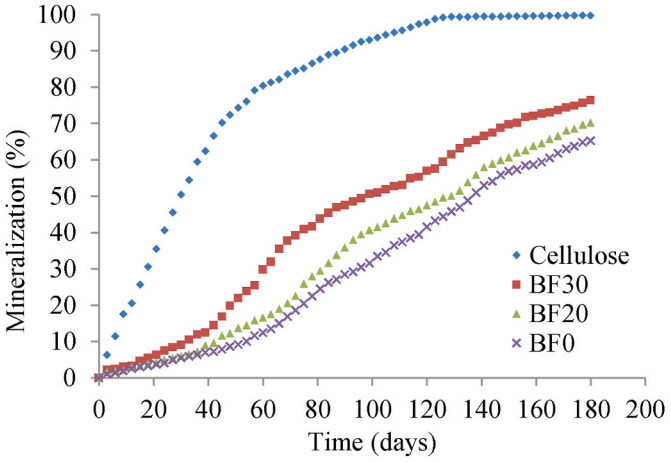

Fig. 15. The mineralization rates of the BFs and cellulose under controlled composting conditions.

Table 2
Thermal properties of the BF30 film before and after hydrolysis.

Sample	T_m (°C)	ΔH_m (J/g)	X_c (%)
Original	112.4	5.78	9.82
HCl medium (3 M, 35 days)	115.0	7.34	12.45
NaOH medium (pH 13, 14 days)	116.4	8.65	14.68
Phosphate buffer (140 days)	113.4	6.40	10.86
Artificial seawater (140 days)	113.2	6.36	10.79

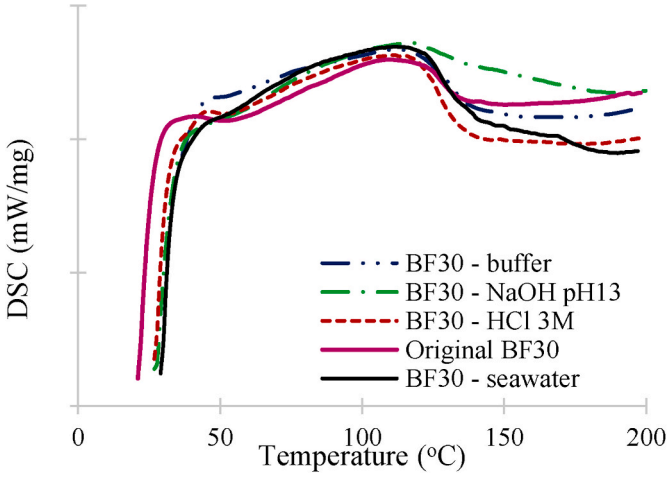

Fig. 14. The DSC curve of the BF30 film before and after hydrolysis.

of the BF0. By this mean, the biodegradation of the BFs was much dependent on the starch content. It also demonstrated that the biodegradation of starch is always better than that of the PBAT. Microorganisms and water were responsible for the α(1–4)- and α(1–6)-glucoside bond cleavages, reducing the chain length of starch, and forming small molecules, CO_2, and H_2O [26]. In addition, water was easily absorbed by starch due to its high hydrophilicity [26].

The biodegradability of the PBAT was deduced from heteroatoms, carbonyl, and aliphatic chains [43,44]. Under hydrolytic and microbiological conditions, the aliphatic BA units (amorphous region) were degraded faster than the aromatic BT units (crystalline area) [45]. The enzymes that were released by the microorganisms in composting accelerated the degradation of BFs. By the standard ASTM D6400, the biodegradable criterion for a potential polymer is more than 60 % [46]. After 180 days of incubation, the biodegradable rates of BF30, BF20, and BF0 were 76.31, 70.14, and 65.25 %, respectively. In previous studies, the biodegradation efficacy of PBAT/cassava starch after 47 days was 38 % [47]. The film of PBAT and reed fiber was accompanied by a biodegradation rate of 11.8 % for 91 days [48]. Muniyasamy et al. (2013) found the biodegradable efficacy of 90 % for PBAT in powders after 190 days and even higher BE, about 100 %, for blends of PBAT with the dried grains [49]. Taken together, the films by blending canna starch and PBAT could be potential materials to replace traditional non-biodegradable plastic products.

4. Conclusions

From this study, the blend PBAT/TPS films have a faster hydrolytic rate than the PBAT film. Its weight loss and mechanical properties showed a tendency in mediums as follows: alkaline > acid > buffer > artificial seawater. In acidic and alkaline solutions, the hydrolysis of the PBAT/TPS films was accelerated as the solution concentration increased. After hydrolysis, there were positional shifts and decreases in the IR peak intensity of the PBAT/TPS films. The SEM images demonstrated that the surface of the PBAT/TPS films after hydrolysis contained holes and pores, especially in the alkaline solution. The melting temperature and crystallinity of the hydrolyzed PBAT/TPS films were higher than those of the original film. Starch could promote the degradation of the PBAT film in the aqueous solution. The complete abiotic hydrolysis of the BF films was 3 M HCl for 14 days or NaOH (pH 13) for 35 days. The biodegradability of the blend films showed an uptrend with increasing the TPS content. The biodegradable rate of the BF30 film after 180 days of incubation in composting was 76.31 %. Collectively, biotic degradation experiments in solid, sewage, or marine water under simulated or real-time exposure conditions are necessary.

Data availability

Data will made available on request.

CRediT authorship contribution statement

Pham Thi Thu Ha: Formal analysis. **Ninh The Son:** Writing – review & editing. **Nguyen Van Khoi:** Supervision. **Pham Thu Trang:** Writing – original draft, Conceptualization. **Nguyen Trung Duc:** Data curation. **Pham Ngoc Anh:** Investigation. **Nguyen Ngoc Linh:** Investigation. **Nguyen Thanh Tung:** Supervision, Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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