



Mechanical Property Deterioration of PBAT/PLA/ENR Mulching Films for the Cultivation of Fruit Trees and Flowers Under Real Field Conditions

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Abstract: The objective of this study is to develop biodegradable mulch films containing PLA and modified natural rubber and study the mechanical behaviour of the biodegradable mulch films containing polylactic acid and modified natural rubber under real field conditions. The present study investigated the effect of different component ratios of the mechanical and biodegradable properties on the bio-based mulching film. Bio-based mulching films were prepared using blown film extrusion with extruder barrel temperature settings ranging between 167 to 170 C and an extruder pressure of 119 bar. The blend composition consisted of Poly (Butylene Adipate-co-Terephthalate) (PBA T), Epoxidized Natural Rubber (ENR), and Polylactic Acid (PLA). The effect of real field conditions on the mechanical properties of the mulching films during the time of cultivation was investigated using a universal testing machine and the pendulum tear method. The mechanical property deterioration of the PLA/ENR mulching films under real field conditions was due to biodegradation caused by biological activity, temperature, and humidity. The results show that the performance of the biodegradable mulching films under actual user conditions for six months was still effective, and the quality of the fruit trees and flowers improved when using the PLA/ENR mulching films.

Keywords: Mulching film, biodegradation, mechanical properties, epoxidized natural rubber, PLA

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I. INTRODUCTION

Polyethylene and polypropylene mulching films have seen general use in agriculture as mulch films. Over the last decade, mulch film consumption obtained from conventional plastics has greatly increased due to the reduction of certain insect pests, moisture conservation, and higher crop yields and quality [1]. In western European markets, agri-related plastic consumption amounts to 700,000 tons/year according to Jean-Claude Garnaud of the International Committee on Plastics in Agriculture [2]. Film applications make up the dominant volume of

this plastic use. An estimation obtained from Applied Market Information (AMI) suggests that the European market use approximately 500,000 tons/year of agricultural films. Other reports reveal that the agricultural film market is growing at about 2.5% per year overall. Due to demands for early fruit and vegetable crops, greenhouse film applications have experienced the fastest growth, while the disposal of used plastic mulch films results in environmental pollution. The total amount of plastic use in agriculture is considerable, and nearly 50% is from the use of films. Approximately 1.2 million tons of agricul-

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tural plastics require disposal each year. Around 26% of agricultural plastics are employed for energetic recovery, and over 600,000 tons end up in landfills [3, 4, 5].

Because of its flexibility, PBAT is an excellent candidate for biodegradable film production and has the potential to reduce the dependence on petrochemical-based plastics and related environmental problems. However, PBAT is much more costly than commercial film as well as when compared to the biodegradable films containing natural rubber developed in this study due to the low cost of natural rubber.

In addition, PLA is also a good candidate for blending with PBAT due to its biodegradability and lower cost, yet its use is limited because of its brittle properties and less flexible chemical chain compared to PBAT.

However, natural rubber is a biodegradable polymer that has high tensile strength, high elongation, and good impact properties. Hence, in this research, natural rubber is used to enhance the flexibility of polymer blends. In addition, natural rubber is one of the most important economic crops produced in Thailand and the country is a top global exporter of natural rubber. The modified natural rubber was fabricated to attain the desired properties. The natural rubber used in this research is Epoxidized Natural Rubber (ENR) with an epoxidation level of 50 percent. A large amount of epoxy-functional group contained in the

polymer chain results in compatibility between polymer phases in the melt blending process.

Additionally, the blending of modified natural rubber is biodegradable mulch film led to lower costs for the biodegradable mulch film which reduced the cost of natural rubber compared to biodegradable bio-based plastics [6]. Meanwhile, some researchers report that blending modified natural rubber with a biodegradable polymer leads to the improvement of various properties.

As previously mentioned, biodegradable mulch films containing PLA and modified natural rubber are derived from renewable materials to replace petroleum-based plastic mulching films. Thailand is the world's top natural rubber producer and exporter according to data obtained from the International Trade Centre. Together, Thailand, Indonesia, and Malaysia make up 70 percent of the world's natural rubber output [7]. A further objective of the present study is therefore to create an opportunity for adding value to natural rubber which can result in high-profit generation and contribute to generating income for Thai citizens.

The objective of this study was to develop biodegradable mulch films containing PLA and modified natural rubber and study the mechanical behavior of the biodegradable mulch films containing polylactic acid and modified natural rubber under real field conditions.

TABLE 1
NAME AND COMPOSITION OF SAMPLES

	PBAT (wt%)	PLA (wt%)	ENR (wt%)	Richnox b215 (phr)	Tinuvin 160 (phr)	Carbon Black (phr)
P811	80	10	10	0.1	1.0	-
P811 - C	80	10	10	0.1	1.0	0.1
P721	70	20	10	0.1	1.0	-
P721 - C	70	20	10	0.1	1.0	0.1

II. MATERIALS AND METHODS

A. Materials

The materials used in this research included PLA (Ingeo 2003D, extrusion grade; NatureWorks, USA) with a density of 1.24 g/cm³ and melting point of 165° and PBAT (ecoflex F Blend C1200; BASF, Germany) with a density of 1.25–1.27 g/cm³ and a melting point of 110–120 °C. ENR containing 50 mol% of epoxidation (ENR50) purchased from Muang Mai Guthrie Public Co. Ltd. (Thailand). Richnox B215 was used as an antioxidant additive and supplied by Salee Industry Public Co., Ltd. Tinuvin

1600 which was used as a UV stabilizer and purchased from BASF (THAI) Ltd.

B. Biodegradable Mulch Film Preparation

1) *Polymer blends composition:* Biodegradable polymer blends of PBAT/PLA/ENR were prepared in two ratios of 80/10/10 and 70/20/10 which are denoted as P811 and P721, respectively, while P811-C and P721-C were mixed with 0.1 phr of carbon black, as shown in Table 1. Richnox B215 was used as an antioxidant additive and supplied by Salee Industry Public Co., Ltd. Tinuvin 1600, UV stabilizer was purchased from BASF (THAI) Ltd.

Richnox b215 and Tinuvin 160 were added into every sequence for 0.1 phr and 1.0 phr, respectively as shown in Table 1.

2) *Polymer blends for preparation:* Polymer pellets were dried in a hot-air oven at 50° for 8 hours. Biodegradable polymers and additives of each ratio were then melt-blending simultaneously by a twin-screw extruder (Brabender Plasti-Corder Extruder PL-2100, Germany) with a temperature profile of 140, 160, 170, and 175° from feed zone to die nozzle at a screw rotation speed of 60 rpm.

3) *Film preparation:* Polymer blend pellets were dried in a hot-air oven at 50° for 8 hours. Polymer pellets were filmed by an extrusion film blowing machine (Lab Tech Engineering Company Ltd.). Three-zone temperature degrees were set at 167°, 169°, and 170° at the die nozzle with a screw speed of 55 rpm and a pressure of 119 bar.

C. Characterization

1) *Tensile testing:* Tensile properties were investigated using a Universal Testing Machine (Instron model 1123) with a crosshead speed of 500 mm/min. Specimens with dimensions of 5 cm (*W*) x 10 cm (*L*) were prepared according to ASTM D882-18.

The tensile properties of the plastic were evaluated

using the following equation:

$$\sigma = \frac{W}{A_o} \quad (1)$$

$$\%EL = \frac{L - L_o}{L_o} \times 100 \quad (2)$$

$$E = \frac{\sigma}{\varepsilon} \quad (3)$$

where, σ is tensile stress, %EL is a percent elongation, *L* is the distance between gauge marks at any time, *L*₀ is the original distance between gauge marks, ε is strain, and *E* is the modulus of elasticity.

2) *Tear resistance:* Tear resistance testing was investigated according to ASTM D1922 - 15 using the pendulum method (Elmendorf Tearing Tester, USA). The blown films were cut into rectangular samples of 76 mm (*W*) and 63 mm (*L*).

3) *Degradation of biodegradable films:* Biodegradability properties were measured by weight change at different times, as shown in Equation 4.

$$\text{Weight loss}(\%) = \frac{(W_i - W_t)}{W_i} \times 100 \quad (4)$$

Note: *W*_i is weight at initiation, *W*_t is weight at the time of investigation.



Fig. 1. Use of biodegradable mulching films for cultivation

TABLE 2
TENSILE YIELD STRESS OF PBAT/PLA/ENR MULCHING FILM

Sample Code	Tensile Strength (MPa)						
	0 M	1 M	2 M	3 M	4 M	5 M	6 M
P811	15.67±1.90	15.32±2.16	14.66±1.57	13.78±1.42	12.91±2.18	12.07±2.16	11.49±1.82
P811-C	15.84±1.41	14.99±0.90	14.09±1.05	13.94±1.79	12.97±1.21	12.10±1.68	11.72±1.69
P721	16.83±1.60	16.18±1.53	15.55±1.51	14.73±1.35	13.12±1.51	12.70±0.88	12.09±1.43
P721-C	16.90±1.24	16.25±0.91	15.24±0.89	14.95±1.19	13.36±1.65	12.83±1.74	12.29±1.84

M : Months after transplanting.

TABLE 3
PERCENT ELONGATION AT BREAK OF PBAT/PLA/ENR MULCHING FILM

Sample Code	Elongation at Break (MPa)						
	0 M	1 M	2 M	3 M	4 M	5 M	6 M
P811	334.83 ± 19.35	328.09 ± 14.82	317.55 ± 14.64	321.71 ± 9.96	308.80 ± 20.06	303.38 ± 7.92	300.93 ± 8.10
P811-C	352.40 ± 28.19	343.04 ± 20.54	335.58 ± 21.41	328.47 ± 21.67	316.05 ± 21.49	309.27 ± 19.35	307.03 ± 10.49
P721	253.71 ± 8.82	250.96 ± 6.21	248.58 ± 2.67	247.35 ± 4.59	238.74 ± 7.49	233.28 ± 10.30	230.82 ± 5.94
P721-C	263.02 ± 9.58	260.56 ± 10.18	255.75 ± 10.16	250.63 ± 9.06	240.04 ± 12.02	237.64 ± 4.08	232.24 ± 12.98

TABLE 4
YOUNG'S MODULUS OF PBAT/PLA/ENR MULCHING FILM

Sample Code	Young's Modulus (MPa)						
	0 M	1 M	2 M	3 M	4 M	5 M	6 M
P811	4.69 ± 0.59	4.68 ± 0.68	4.57 ± 0.54	4.34 ± 0.45	4.16 ± 0.42	3.99 ± 0.78	3.83 ± 0.70
P811-C	4.53 ± 0.66	4.38 ± 0.25	4.22 ± 0.51	4.27 ± 0.67	4.12 ± 0.41	3.91 ± 0.43	3.82 ± 0.55
P721	6.63 ± 0.54	6.46 ± 0.72	6.26 ± 0.59	5.96 ± 0.50	5.48 ± 0.49	5.45 ± 0.37	5.23 ± 0.57
P721-C	6.43 ± 0.50	6.25 ± 0.44	5.97 ± 0.45	5.98 ± 0.55	5.59 ± 0.81	5.40 ± 0.77	5.31 ± 0.83

III. RESULTS AND DISCUSSION

The tensile properties of the biodegradable mulch films (P811, P811-C, P721, and P721-C) are shown in Tables 2, 3, and 4. These tables reveal the tensile yield stress, elongation at break, and Young's modulus of the biodegradable films.

The results show that PBAT/PLA/ENR blends tend to increase tensile stress and Young's modulus, but a decrease in elongation with higher a PLA content ratio from 10 to 20 wt% due to the brittleness of PLA.

In this study, PBAT is the main component of the PBAT/PLA/ENR blends with 70-80 wt% of PBAT content. Therefore, PBAT is the matrix phase and the other

polymers are dispersed phases, including additive particles of carbon black, antioxidation agent, and UV stabilizer.

The increased PLA content up to 20 wt% in the blends resulted in increased tensile yield stress and Young's modulus which is higher than the blends which contained 10 wt% PLA content. Meanwhile, elongation at the break dramatically decreased from $334.83 \pm 19.35\%$ (P811) to $253.71 \pm 8.82\%$ (P721). The results indicate that the decreased tensile properties at higher PLA content is due to weak interfacial interactions, resulting in incompatibility between PBAT and PLA [8]. Since the PLA structure is stiffer than PBAT, this indicates that the PLA became

more brittle than PBAT which has a long flexible chemical chain [9].

The biodegradable films with carbon black, P811-C, and P721-C showed higher tensile yield stress and percent elongation at break than the other films of unfilled carbon black (P811 and P721) because carbon black particles behave like a reinforcing agent in the polymer matrix. The addition of carbon black particles in the PBAT/PLA blends resulted in greater crystallinity which had the effect of increasing tensile strength in order for the carbon black particles to act as an initiating agent in the system to develop the crystallinity of the blends [10, 11, 12].

After mulching in real cultivation conditions, the results of tensile tests were recorded every month for six months. The results reveal that the mechanical properties such as the tensile yield stress, % elongation, and Young's modulus of all the samples tended to decrease with increased mulching time from 0-6 months. The biodegradation of the biodegradable polymer blends is behind the loss of strength since the chemical chains broke down and their performance decreased, resulting in them becoming small pieces, fragments, and changing into other forms such as methane gas, carbon dioxide gas, or water by bio-organisms living in the environment [13, 14].

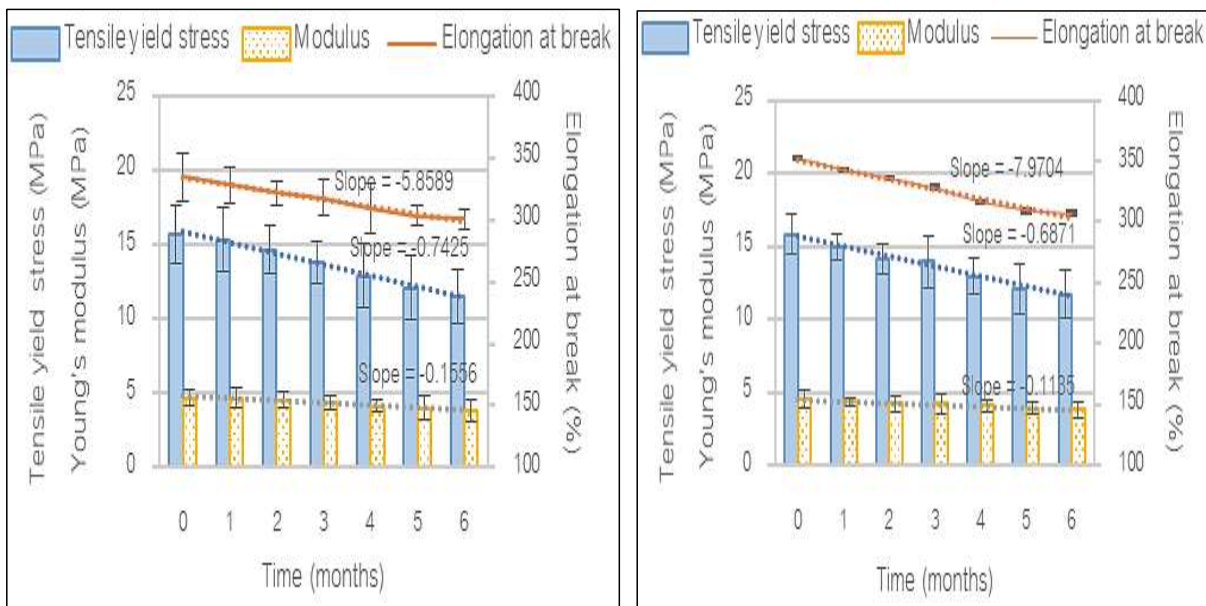


Fig. 2. Tensile properties of the biodegradable polymer blend mulching film of (a) P811 and (b) P811-C as a function of degradation time

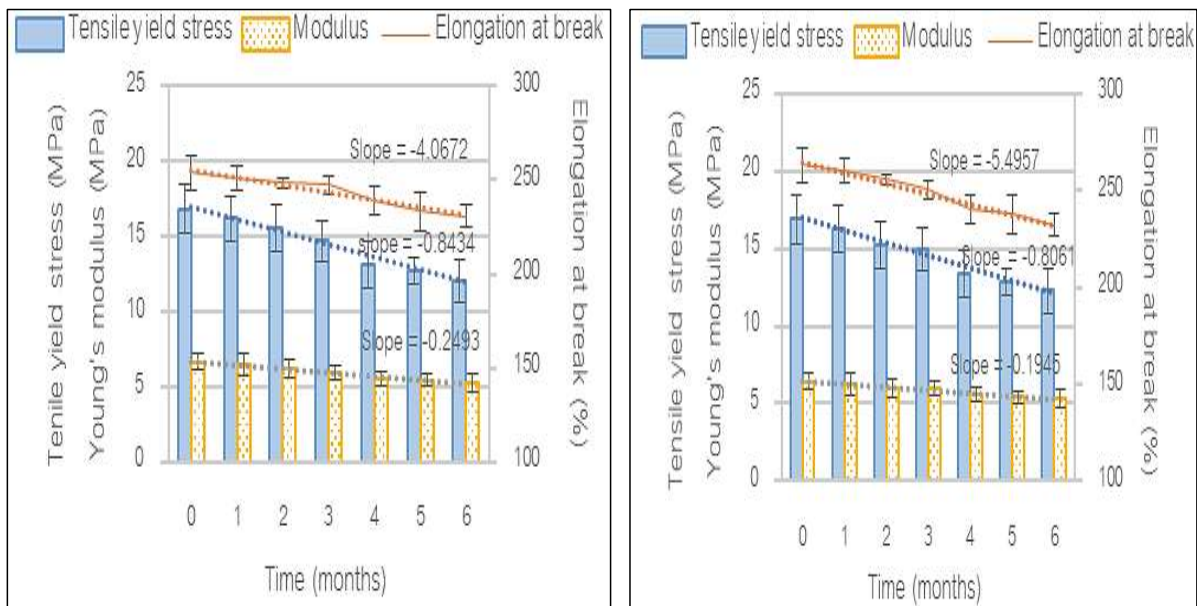


Fig. 3. Tensile properties of the biodegradable polymer blend mulching film of (a) P721 and (b) P721-C as a function of degradation time.

Fig. 2 and Fig. 3 show the tensile properties of the biodegradable films. These results reveal that the tensile properties decreased with increased mulching periods. Furthermore, the graphs clearly demonstrate the reduced trend line of decreasing mechanical performance with increasing mulching period. The graphs represent the tensile performance at various mulching times using the biodegradable films which generate trend lines that explain the reduction rate of the film performance.

TABLE 5
DERIVED SLOPE OBTAINED FROM THE GRAPH PLOT
OF TENSILE PROPERTIES AS A FUNCTION OF
DEGRADATION TIME

Samples	Trendline Slope		
	Tensile Yield Stress	% at Break	Modulus
P811	-0.7425	-5.8589	-0.1556
P811-C	-0.6871	-7.9704	-0.1135
P721	-0.8434	-4.0672	-0.2493
P721-C	-0.8061	-5.4957	-0.1945

The slope of the trendlines between tensile properties versus time represents the performance loss rate of the films. Comparing between the formulas, P721 had the highest reduction rate of tensile strength performance followed by P721-C, P811, and P811-C, respectively. The trendlines of Young's modulus show similar trends as tensile strength. Meanwhile, P811-C shows the highest reduction rate of elongation performance, followed by P811, P721-C, and P721, respectively. However, films containing carbon black exhibited lower reduction rates compared to unfilled films (P721-C and P811-C) because carbon black is a photo stabilizing agent that can absorb ultraviolet energy and slow the photo-oxidation mechanism [15]. Photodegradation is one of the main causes of the degradation of mulching film, in which UV radiation absorbed from sunlight causes photooxidation. UV radiation has the effect of debonding and even crosslinking polymer chain structures in the degradation processes of PBAT [16]. Exposure to sunlight with moisture in the air and soil also causes changes in the PLA chain structure through the hydrolysis chain scission mechanism [17]. Therefore, losing flexibility by chain scission and crosslinking mechanism are shown through the steeper, elongated reduction slope of the films containing 80 wt% of PBAT.

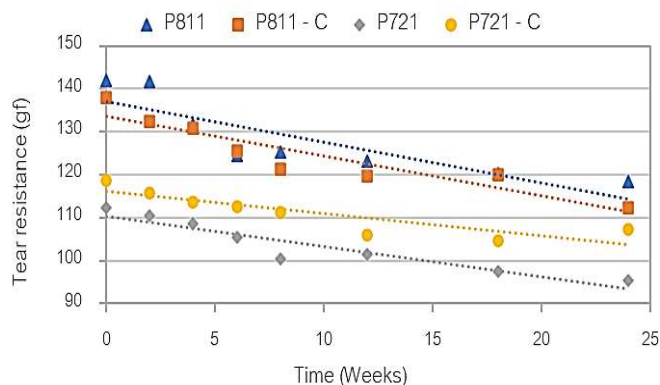


Fig. 4. Tear resistance of the biodegradable polymer blend mulching films P811, P811-C, P721, and P721-C as a function of degradation time

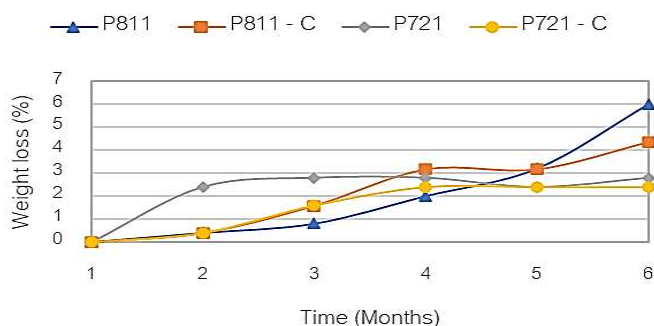


Fig. 5. Percentage of weight loss of the biodegradable polymer blend mulching films P811, P811-C, P721, and P721-C as a function of degradation time

Fig. 4 shows the tear resistance of the biodegradable films as a function of time. The results reveal that biodegradable films containing 80% of PBAT have higher performance than the films containing 70% of PBAT content due to the higher toughness of biodegradable films containing 80% of PBAT [18].

The degradability of biodegradable films under cultivation conditions, in which the mulching films are exposed to light, weather, and bio-organisms in the environment. Hence, direct contact between the mulching film and UV radiation, air, heat, and moisture are the main factors of polymer chain scission [19]. The degradation of biodegradable films generally occurs through photo-oxidative, thermal-oxidative, and hydrolysis mechanisms [20].

Irradiation firstly attacks the interface between crystalline and amorphous which operate via the radical mechanism with excessive oxygen in the environment to generate photo-oxidation [21, 22]. As is widely known, biodegradable polymers which have polar part can absorb much more moisture than conventional plastics through the functional group on the chemical chain, such as carbonyl group by the hydrogen bond [23].

For mulch films, it is used as a covering layer on outdoor land, meaning that under the film there is an increase of heat greater than the surrounding atmosphere, which leads to the hydrolysis mechanism [23]. Hydrolysis of ester linkage bonds in the bone structure of biodegradable polymer occurs at temperatures above 30° with moisture [24]. Furthermore, UV radiation could enhance the degradation rate compared to films untreated with UV irradiation [25]. These degradations were investigated by the overall weight loss percentage over a period of six months. As shown in Fig. 5, the biodegradable blends P721, P721-C, P811, and P811-C tended to decrease with increased time of use due to the biodegradability of the polymer blends. Polymer blend films containing carbon black exhibited lower weight loss percentages compared to the films without carbon black due to the UV absorption ability of carbon black and the slower photo-oxidation reaction of the polymer. The P811 sample had higher amounts of PBAT than P721 and showed a higher weight loss percentage than P721 at the end of the investigation. This indicates that biological decomposition in the structure of PBAT at the surface and significant chemical hydrolysis takes place within the co-polyesters [26].

IV. CONCLUSION AND RECOMMENDATIONS

The PBAT/PLA/ENR mulch films containing carbon black were prepared in this study. The P811-C sample showed the highest mechanical property performance, while the P721 sample showed lower performance. In addition, the blending of natural rubber in the blend films led to enhanced biodegradability and properties. This study encourages scholar to extend this study and investigate further the issues concerning.

Declaration of Conflicting Interests

It is an original work carried out by equal contribution of all the authors. The authors declare that there is no conflict of interest.

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