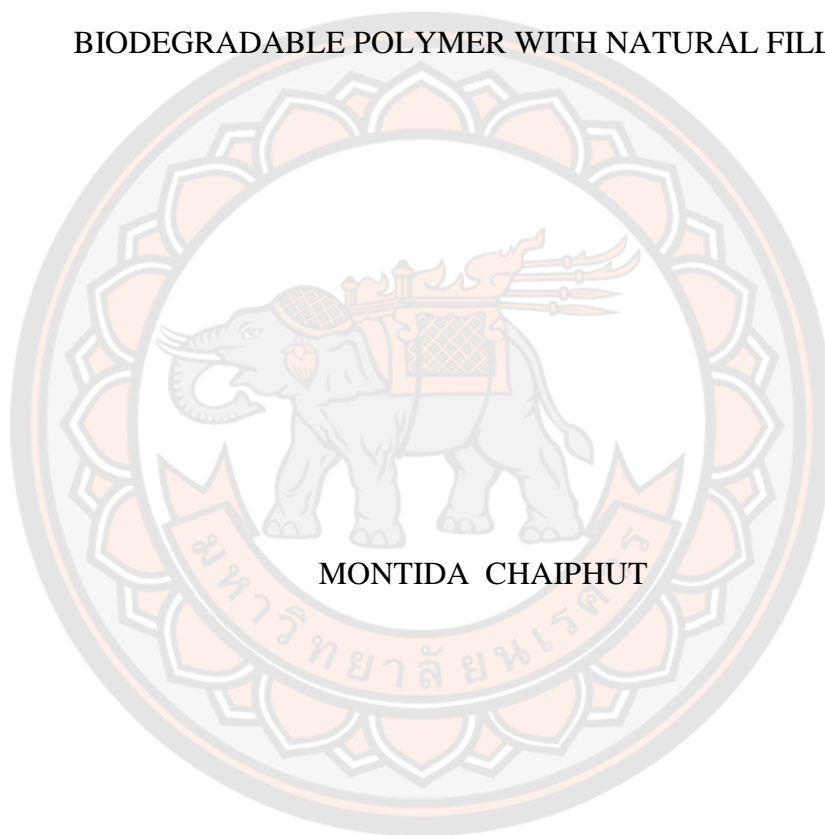




PREPARATION AND CHARACTERIZATION OF MULCH FILM FROM  
BIODEGRADABLE POLYMER WITH NATURAL FILLERS



MONTIDA CHAIPHUT

A Thesis Submitted to the Graduate School of Naresuan University  
in Partial Fulfillment of the Requirements  
for the Master of Science in (Industrial Chemistry)

2020

Copyright by Naresuan University

PREPARATION AND CHARACTERIZATION OF MULCH FILM FROM  
BIODEGRADABLE POLYMER WITH NATURAL FILLERS



A Thesis Submitted to the Graduate School of Naresuan University  
in Partial Fulfillment of the Requirements  
for the Master of Science in (Industrial Chemistry)

2020

Copyright by Naresuan University

Thesis entitled "Preparation and characterization of mulch film from biodegradable polymer with natural fillers"

By MONTIDA CHAIPHUT

has been approved by the Graduate School as partial fulfillment of the requirements for the Master of Science in Industrial Chemistry of Naresuan University

**Oral Defense Committee**

..... Chair

(Assistant Professor Sirisart Ouajai)

..... Advisor

(Assistant Professor Sararat Mahasaranon)

..... Co Advisor

(Assistant Professor Nungruthai Suphrom)

..... Internal Examiner

(Assistant Professor Supatra Pratumshat)

**Approved**

.....  
(Professor Paisarn Muneesawang)

Dean of the Graduate School

<b>Title</b>	PREPARATION AND CHARACTERIZATION OF MULCH FILM FROM BIODEGRADABLE POLYMER WITH NATURAL FILLERS
<b>Author</b>	MONTIDA CHAIPHUT
<b>Advisor</b>	Assistant Professor Sararat Mahasaranon
<b>Co-Advisor</b>	Assistant Professor Nungruthai Suphrom
<b>Academic Paper</b>	Thesis M.S. in Industrial Chemistry, Naresuan University, 2020
<b>Keywords</b>	Poly (lactic acid) Polybutylene succinate natural fillers Bio-composite film

### **ABSTRACT**

This research has been interested in developing and producing the agriculture product as mulch film from biodegradable polymer incorporated with natural fillers. To investigate the effect of natural fillers on the physical properties, mechanical properties, degradability, and field test. The biodegradable polymers were used in this work including poly(lactic acid); PLA and polybutylene succinate; PBS. The natural fillers were used early immature ginger (EG), mature ginger (MG), and lemongrass leaf (LE) with approximately 45  $\mu\text{m}$  of particle size. All the natural fillers are Thai herbal and easier to found in the country. First section, the natural fillers were compounded with PLA and then the good natural filler was chosen for improving the properties of bio-composite film. The addition of natural filler affects the mechanical of biocomposite film was reduced due to the continuous phase was reduced by natural fillers. Especially, the addition of ginger powder causing generated more agglomerate in the polymer matrix. The physical properties were found the addition of natural filler can be improved the colorlessness of PLA

that help to reduce the UV-light transmit through out the biocomposite film. The addition of LE powder can absorb the highest UV-light that shows the UV-light can transmit around 1-2%. This section can be summarized, the use of LE powder is good natural filler for improving the light barrier. However, the selection of only PLA as a polymer matrix probably unsuitable due to PLA has hard and brittle. This research has been interested to used PBS for improve the flexibility of polymer matrix. The addition of PBS into the PLA matrix was developed the flexibility and strength of the PLA, which increasing the tensile strength and %elongation of the biopolymer film. LE powder was used for natural filler to prevent UV-light transmittance. Because the requirement of mulch film must be loss of UV-light transmittance for manage the weed generation. The addition of LE powder can be reduced the UV-light transmittance that absorbs the UV-light high to 90%. The biocomposite film was carried on the field under environment for 2 months, which used the real condition for agriculture crop. To investigate the performance of LE powder on the mechanical and physical properties (color parameter and %UV transmittance). The color of biocomposite film was changed to pale color with greater field time which believes that cause from the reaction of compounds in the LE powder changes. While in after 2 months, the biocomposite film also absorbs the UV-light at a high level (~90%). The changing of color is hardly affecting the UV-light transmittance. The mechanical properties of biocomposite film after 2 months were decreased trend but less different value when compared with start value. The reduction of mechanical properties probably the chain scission from water, microbial, and sunlight. The addition of LE powder into biocomposite film has the excellent advantage that prevent weed

growth, reduce toxic because all material has non-toxic chemical and biodegradable. Especially, the use of natural fillers as herbal and plants in-country can help to increase the value of these plant and steam revenue to farmers, and the community.



## ACKNOWLEDGEMENTS

This thesis studied about the preparation and characterization of the biocomposite film based on biopolymer and natural fillers for producing the mulch film.

First of all, I would like to express my special thanks to my advisor, Assistant Professor Dr. Sararat Mahasaranon, who gave me the opportunity to do this research, and thankfulness for supporting me in everything. Secondly, I would like to thank co-advisors; Assistant Professor Dr. Nungruthai Suphrom, Assistant Professor Dr. Supatra Pratumshat as an examiner, and Assistant Professor Dr. Gareth Ross for reviews my manuscript and help throughout my study and research. Special thanks to Assistant Professor Sirisart Ouajai for his suggestions.

The authors would like to thank the Department of Chemistry, Faculty of Science, Naresuan University, and Science Lab Center for support and training about using the instrument. In addition, I was fortunate to have the opportunity and scholarships of the Development and Promotion of Science and Technology Talents Project (DPST) to support the grant.

Finally, I do not forget to thank the Biopolymer group and special thanks to Ms. Amonrut Waisarikit for helping me in everything. It is very kind of you. Moreover, my family in need with treatment and support me. I could not have done it without everyone. Thank you.

MONTIDA CHAIPHUT

## TABLE OF CONTENTS

	<b>Page</b>
ABSTRACT.....	C
ACKNOWLEDGEMENTS.....	F
TABLE OF CONTENTS.....	G
LIST OF TABLES.....	J
LIST OF FIGURES.....	L
CHAPTER I INTRODUCTION.....	1
Background and significant of the study.....	1
Purposes of the study.....	3
Scope of the study.....	3
CHAPTER II LITERATURE REVIEWS.....	5
Mulch film.....	5
1. The mulch film in commercial.....	6
Manufactory of mulch film.....	10
1. Extrusion.....	10
2. Blow film extrusion.....	13
Polymer blend and composite.....	14
1. Polymer blend.....	14
2. Polymer composite.....	17
Bioplastic.....	20
1. Poly(lactic acid).....	20
2. Polybutylene succinate.....	24
Natural fillers.....	26
1. Ginger.....	27
3. Lemongrass.....	32
CHAPTER III RESEARCH METHODOLOGY.....	35



Raw materials .....	35
Equipment.....	35
Instrument .....	36
Methodology and characterization technique .....	36
1. Natural fillers.....	36
2. The bio-composite film from poly(lactic acid) incorporated with natural fillers.....	37
3. The bio-polymer blend film from poly(lactic acid) incorporated with polybutylene succinate .....	42
4. The bio-composite film from the bio-polymer blend incorporated with lemongrass leaf powder.....	45
CHAPTER IV RESULTS AND DISCUSSION.....	49
The characterization of natural fillers .....	49
1. The chemical properties of the natural fillers.....	50
2. The morphology of the surface natural fillers .....	51
3. Fiber length of natural fillers.....	54
The effect of each natural filler in the bio-composite base on PLA matrix.....	55
1. Bio-composite compound from PLA and natural fillers .....	55
2. Bio-composite film from PLA and natural fillers .....	61
The effect of polybutylene succinate (PBS) in the bio-polymer blend base on PLA matrix.....	92
1. Bio-polymer blend compound.....	92
2. Bio-polymer blend film .....	96
The effect of polybutylene succinate (PBS) and natural filler as lemongrass leaf powder (LE) in the bio-composite base on PLA matrix.....	107
1. Bio-composite compound from PLA, PBS, and LE powder .....	108
2. Bio-composite film from PLA, PBS, and LE powder.....	112
CHAPTER V CONCLUSIONS .....	144
Conclusions.....	144
Suggestion.....	147
REFERENCES .....	148

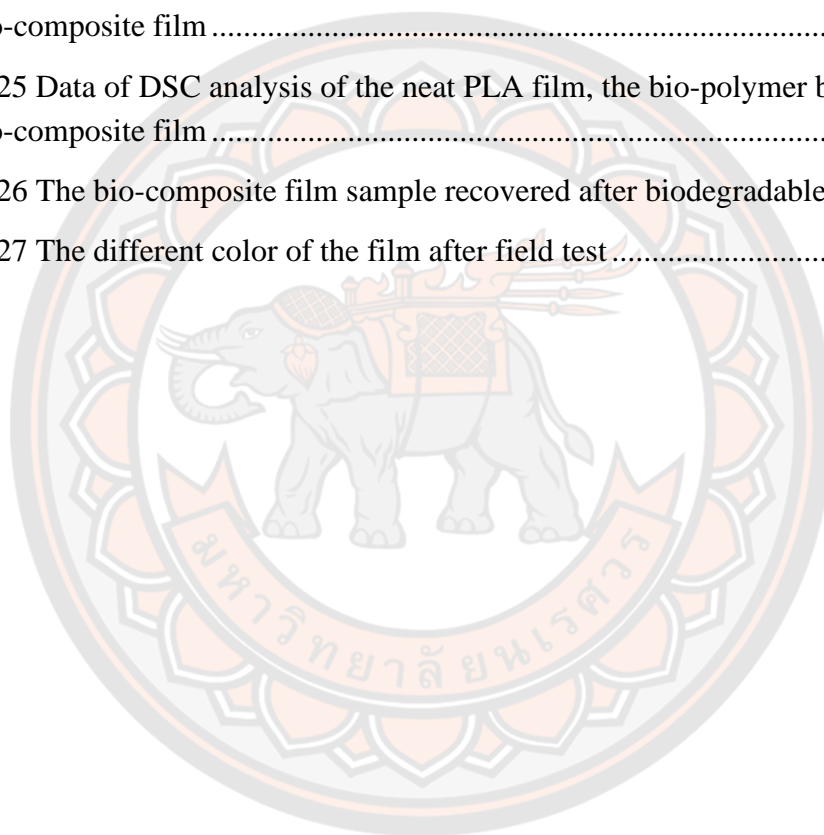
BIOGRAPHY .....153



## LIST OF TABLES

	<b>Page</b>
Table 1 The effect of mulch film on the agricultural crops (11) .....	9
Table 2 The summary of twin-screw rotations (23).....	13
Table 3 Compositions of different maturation stages of ginger .....	31
Table 4 The ratio of the bio-composite film between poly(lactic acid) and each natural fillers .....	38
Table 5 The ratio of the bio-polymer blend film between poly(lactic acid) and various contents polybutylene succinate .....	43
Table 6 The ratio of the bio-composite film between poly(lactic acid) and polybutylene succinate with various the content of lemongrass leaf powder.....	46
Table 7 The morphology of natural fillers .....	53
Table 8 The acronym of bio-composite compound and film.....	55
Table 9 FT-IR spectrum interpretation of natural fillers and the bio-composite film .	63
Table 10 Surface morphology and cross-section of the neat PLA film after carried the tensile testing .....	66
Table 11 Surface morphology and cross-section of EG film after carried the tensile testing.....	67
Table 12 Surface morphology and cross-section of MG film after carried the tensile testing.....	70
Table 13 Surface morphology and cross-section of LE film after carried the tensile testing.....	73
Table 14 The color parameter of the neat PLA film and the bio-composite film.....	83
Table 15 %UV-transmittance of the neat PLA film and the bio-composite film .....	86
Table 16 The bio-composite film sample recovered after biodegradable in soil.....	89
Table 17 The acronym of bio-polymer blend compound and film.....	92
Table 18 Data of DSC analysis of the neat PLA film, the neat PBS film, and the bio-polymer blend film.....	106
Table 19 The acronym of the bio-composite with PBS and various content of lemongrass leaf powder .....	108

Table 20 Cross-section morphology of the neat PLA compound and the bio-composite compound.....	110
Table 21 FT-IR data of natural fillers and the bio-composite film.....	114
Table 22 Surface Morphology and Cross-section of the bio-composite film after tensile testing .....	120
Table 23 The color parameter of the neat PLA film, the bio-polymer blend film, and the bio-composite film .....	127
Table 24 %UV-transmittance of the neat PLA film, the bio-polymer blend film, and the bio-composite film .....	129
Table 25 Data of DSC analysis of the neat PLA film, the bio-polymer blend film, and the bio-composite film .....	132
Table 26 The bio-composite film sample recovered after biodegradable in soil.....	134
Table 27 The different color of the film after field test.....	137



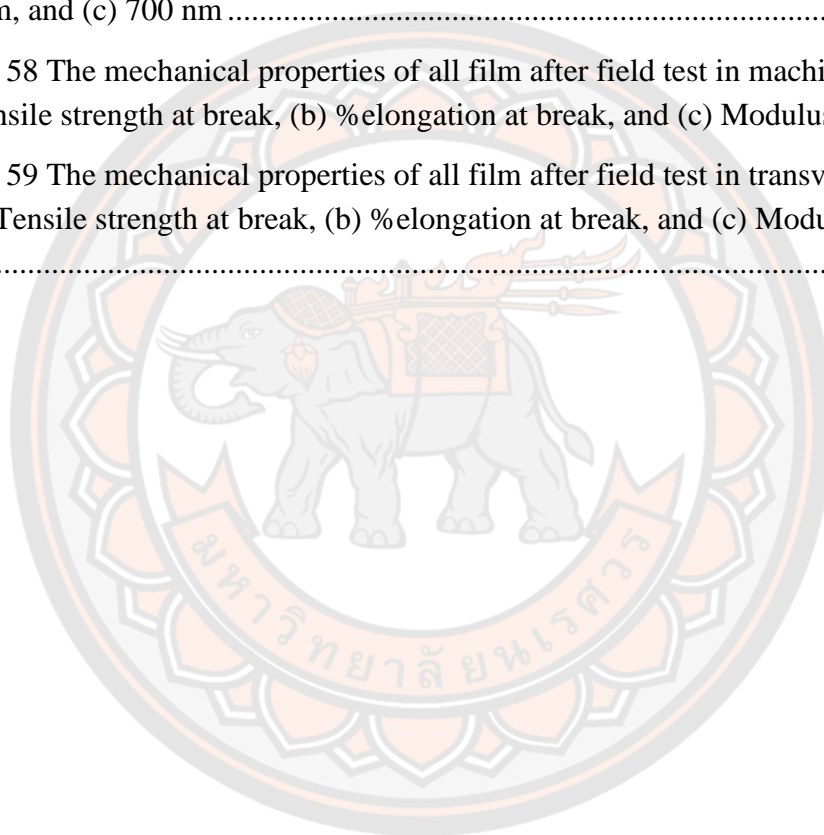
## LIST OF FIGURES

	<b>Page</b>
Figure 1 The black mulch film (12).....	7
Figure 2 The clear mulch film (14).....	7
Figure 3 The white on black and silver reflective mulch film (15).....	8
Figure 4 The extruder screw zone (20).....	11
Figure 5 Single screw extruder (20).....	12
Figure 6 The rotation of twin-screw extruder (22).....	13
Figure 7 The blow film extrusion (24).....	14
Figure 8 Solution casting process.....	16
Figure 9 Batch blending process.....	16
Figure 10 Continuous blending process (28).....	17
Figure 11 The fibrous orientation in the polymer matrix (a) Random orientation of particle, (b) Preferred orientation of discontinuous fibrous, (c) Random orientation of discontinuous fibrous, (d) Uniaxial orientation of continuous fibrous, (e) biaxial orientation of continuous fibrous, and (f) multi-direction orientation of continuous fibrous (30).....	19
Figure 12 Classification of the composite materials (29).....	19
Figure 13 Stereochemistry of (a) PLLA, (b) PDLA, and (c) monomer of PLA (7)....	21
Figure 14 The polymerization of PLA (32).....	21
Figure 15 Hydrolysis degradation mechanism of PLA (34).....	22
Figure 16 Photodegradation mechanism of PLA (35).....	23
Figure 17 Chemical structure monomer of PBS (7).....	24
Figure 18 Enzymatic degradation Mechanism of PBS (38).....	25
Figure 19 The chemical structures of active components from ginger; (a) 6-gingerol, (b) 6-shogaol, and (c) 6-paradol (45).....	28
Figure 20 The appearance of (a) peel ginger and (b) the cross-section of early immature ginger (46).....	28
Figure 21 The chemical structure of curcuminoid compound as (a) curcumin, (b) demethoxycurcumin, and (c) 6-dehydrogerdione (44).....	29

Figure 22 The appearance of (a) peel ginger and (b) the cross-section of mature ginger (46).....	30
Figure 23 Lemongrass ( <i>Cymbopogon citratus</i> ) .....	32
Figure 24 Chemical compound of lemongrass (a) citral a (geranial) and (b) citral b (neral) (53) .....	33
Figure 25 Preparation of the sample of the bio-composite compound and bio-composite film for SEM analysis.....	40
Figure 26 FT-IR spectra of (a) early immature ginger powder, (b) mature ginger powder, and (c) lemongrass leaf powder .....	51
Figure 27 The fiber length of each natural fillers .....	54
Figure 28 Cross-section morphology of PLA compound at magnification (a) 200X, and (b) 1000X .....	56
Figure 29 Cross-section morphology of bio-composite compound incorporated with early immature ginger powder at magnification (a) 200X, and (b) 1000X .....	57
Figure 30 Cross-section morphology of bio-composite compound incorporated with mature ginger powder at magnification (a) 200X, and (b) 1000X .....	58
Figure 31 Cross-section morphology of bio-composite compound incorporated with lemongrass leaf powder at magnification (a) 200X, and (b) 1000X.....	59
Figure 32 Melt flow index of bio-composite compound incorporated with each natural fillers and different content of natural fillers .....	61
Figure 33 FT-IR spectrum of natural fillers as (a) early immature ginger powder, (b) mature ginger powder, (c) lemongrass leaf powder and the bio-composite film as (d) neat PLA film, (e) EG film, (f) MG film, and (g) LE film .....	64
Figure 34 The mechanical properties of bio-composite film incorporated with EG powder as (a) Tensile strength at break, (b) %Elongation at break, and (c) Modulus at break.....	76
Figure 35 The mechanical properties of bio-composite film incorporated with MG powder as (a) Tensile strength at break, (b) %Elongation at break, and (c) Modulus at break.....	77
Figure 36 The mechanical properties of bio-composite film incorporated with LE powder as (a) Tensile strength at break, (b) %Elongation at break, and (c) Modulus at break.....	78
Figure 37 Water vapor transmission (WVTR) of the neat PLA film and the bio-composite film incorporated with the different types and contents of natural filler....	80

Figure 38 Biodegradability of the neat PLA film and the bio-composite film incorporated with (a) MG powder, (b) EG powder, and (c) LE powder .....	91
Figure 39 cross-section morphology of the neat PLA compound at magnification (a) 200X, and (b) 1000X .....	93
Figure 40 Cross-section morphology of the bio-polymer blend based on PLA with 10% w/w of PBS content at magnification (a) 200X, and (b) 1000X .....	94
Figure 41 Cross-section morphology of the bio-polymer blend based on PLA with 20% w/w of PBS content at magnification (a) 200X, and (b) 1000X .....	94
Figure 42 Melt flow index of the bio-polymer blend based on PLA with varies content of PBS .....	96
Figure 43 The cross-section morphology of the neat PLA film at magnification 1000X .....	97
Figure 44 The cross-section morphology of the L90_B10 film at magnification 1000X .....	97
Figure 45 The cross-section morphology of the L80_B20 film at magnification 1000X .....	98
Figure 46 XRD pattern of (a) neat PLA film, (b) PBS film, (c) L90_B10, and (d) L80_B20 .....	100
Figure 47 The mechanical properties of the neat PLA film and the bio-polymer blend film as (a) tensile strength at break, (b) %elongation at break, and (c) modulus at break.....	103
Figure 48 DSC thermograms of the neat PLA film, the neat PBS film, and the bio-polymer blend film.....	106
Figure 49 Melt flow index (MFI) of the neat PLA compound and the bio-composite compound.....	112
Figure 50 FT-IR spectrum of (a) lemongrass leaf powder, (b) neat PLA film, (c) neat PBS film, (d) bio-polymer blend film, and (e) the bio-composite film (B10_LE)....	115
Figure 51 XRD pattern of (a) LE powder (b) B10_LE2.5, (c) B10_LE5.0, (d) B10_LE7.5, and (e) B10_LE10.0 .....	117
Figure 52 Mechanical properties of the bio-composite film as (a) tensile strength at break, (b) %elongation at break, and (c) Modulus at break.....	123
Figure 53 Water vapor transmission (WVTR) of the neat PLA film, the neat PBS film, the bio-polymer blend film, and the bio-composite film with various contents of LE powder.....	125

Figure 54 DSC thermograms of the neat PLA film, the bio-polymer blend film, and the bio-composite film in second heating .....	131
Figure 55 Biodegradability of the neat PLA film, the bio-polymer blend film, and the bio-composite film .....	135
Figure 56 The color parameter (a) L*, (b) a*, and (c) b* values of the film after field test.....	138
Figure 57 Percentage of UV-transmittance of the neat PLA film, the bio-polymer blend film, and the bio-composite film after field test in wavelength at (a) 315 nm, (b) 400 nm, and (c) 700 nm .....	140
Figure 58 The mechanical properties of all film after field test in machine direction as (a) Tensile strength at break, (b) %elongation at break, and (c) Modulus at break...	142
Figure 59 The mechanical properties of all film after field test in transverse direction as (a) Tensile strength at break, (b) %elongation at break, and (c) Modulus at break .....	143





# CHAPTER I

## INTRODUCTION

### **Background and significant of the study**

Agricultures and food have a high growth market in tripled between 1960 and 2015. The forecast has the tendency to increase the market size. The world's population has increasing grow more than 10 billion in 2050 that caused to control of the diet was limited because the area of agriculture has limited availability. (1) The short cycle plants are one choice that respond for requirement of human due to it can get the product all season. The crops of short cycle plants can save times and it can crop in a limitation area. Moreover, the crops of short cycle plant can be easier to steam high revenue for the economic countries. Therefore, the agricultural product must be control and attend to produce high quality and high yield. The plastic mulch film has become used in agriculture which covers the soil that can improve the quality of the crop and increased the yield of the agriculture product. (2) The mulch film used for controlling the crop which can control the soil temperature, keep the moisture, prevent weed growth, protect the agriculture product contact with the soil. Furthermore, the use of plastic mulch film can reduce the herbicide and reduce the toxic chemical released from the herbicide to the environment. (3) However, the use of plastic mulch film affected to the environment. The expansion of the plastic mulch film can be a cause of the rapidly increasing agriculture waste. In 2010, the European plastic industry has produced the plastic more than 57 million tons which have 21.5% of the world's production. Around 5% was to produce agricultural plastic with approximately 2.8 million tons. The increasing of the agricultural plastic production cause promising the agricultural plastic wastes has rapid growing as well. In 2007, the European has generated the agriculture plastic waste more than 700,000 tons per year and promising to growth up. (4)

Generally, the plastic mulch film was produced from synthetic materials such as polyethylene (PE) and low-density polyethylene (LDPE). These materials were performed by the crude oil refining process. The burning process of crude oil that

emission the pollution to the atmosphere more than a billion tons per year. The side effects cause many toxic gases, greenhouse gases that causing global warming and climate change. (1, 3) The PE mulch film is a non-biodegradable material. After harvesting the crop, the PE mulch film will become agricultural waste. The PE mulch film is possible to fragment and become microplastic. The fragmentation products of the PE mulch film can remain in the soil and the groundwater that can release the toxic chemical such as phthalate as a plasticizer, inorganic pigment, and the chemical from herbicide and pesticide. (5) Moreover, the mulch film after removed from the crop has non-recycling because the mulch film was contaminated with the herbicide and pesticide. The mulch film may be contaminated with the disease if lead to recycling affects the crop was fallen and die. (5, 6) Nowadays, bio-polymer has been interested to become the biodegradation mulch film production. Poly(lactic acid); PLA is the bio-polymer made from the renewable resource which can be biodegradable in environment. However, the limitation for the application of PLA has hard and brittle properties and colorlessness. Furthermore, PLA has a rather high production cost than that of PE. (7) The limitation of PLA causing this research has interested to improve the flexibility of PLA. Moreover, the main properties of mulch film must be a light barrier, but the PLA has colorlessness. This research has been interested to improve the colorlessness of PLA by adding the natural filler.

The purpose of this work has been to produce and characterize the mulch film from the bio-polymer incorporated with the natural fillers. The mulch film manufactory was produced by the plant machine include a twin-screw extruder and blow film extruder. The bio-polymer film used in this work consists of poly(lactic acid) and polybutylene succinate (PBS). PBS was added for improving the flexibility of the PLA. Moreover, this study also interested to improve the colorlessness and reduce the UV-light transmittance of the bio-composite film by adding natural fillers. Ginger powder and lemongrass leaf powder as two natural fillers were added into the bio-composite films, which has improved the property to reduce %UV-transmittance passes the bio-composite film to the soil that may prevent weed growth. Moreover, the addition of the natural filler can be improved the biodegradability of the bio-composite film, which the bio-composite film has degraded that become to carbon dioxide and water. The selection of using the natural filler for adding into the bio-

composite film were non-toxic, biodegradation and non-releasing the chemical toxic to the environment. Especially, the ginger and lemongrass as natural fillers are Thai herbal that can found in the country thus sufficient for the mulch film production process. Moreover, the use of these plants can steam revenue to farmers and the economy in the countries.

### **Purposes of the study**

1. To produce the mulch film from the bio-composite film incorporated with natural fillers by a twin-screw extruder and the blow film extruder
2. To study and find the natural filler for improving the properties of the bio-composite film
3. To study the appropriate ratio between PLA and PBS that improved the flexibility of the bio-composite film.
4. To study and improve the colorlessness and reduce %UV transmittance of the bio-polymer by adding the natural filler.

### **Scope of the study**

This research studied and investigated the natural fillers, the bio-composite compound, and the bio-composite film. The studies were divided into 4 sections.

1. To study the characteristics and fiber length of the natural filler to examine the physical and chemical properties which are morphology and functional group of the natural filler, respectively.
2. To study the effect of the natural fillers (ginger powder and lemongrass leaf powder) in the bio-composite film. Effect on chemical properties (FT-IR), physical properties (color parameter, %UV transmittance, water vapor permeability), mechanical properties (tensile testing), and biodegradability were studied. After, appropriate natural filler was chosen to improve the properties of the bio-composite film.
3. To study the effect of content of PBS adding into the PLA film, which select the suitable content of PBS to improve the flexibility of the bio-polymer film. Moreover, effect on the physical properties (morphology), chemical properties

(XRD), mechanical properties (Tensile testing), thermal properties (DSC) were examined.

4. To study and discuss the effect of the lemongrass leaf powder as natural filler and content of PBS in the bio-composite film. Chemical properties (FT-IR, XRD), physical properties (color parameter, %UV transmittance, water vapor permeability), mechanical properties (tensile testing), thermal properties (DSC), biodegradability, and field test, were examined, respectively.



## CHAPTER II

### LITERATURE REVIEWS

The literature reviews in this research have been interested to study about the mulch film, the manufactory of mulch film, and the mulch film in the commercial. Furthermore, this research has also been interested about the bio-polymer from renewable resource and natural fillers. The details of each topic were illustrated as following.

#### **Mulch film**

The mulch film has been used in commercials for the vegetable crops since the 1960s. The most color of mulch film are black and clear. However, color of the mulch film influenced the soil and crop have differently in the growing of plants. (2) The mulch film is the application of plastic film used to cover agricultural plots due to it has convenient and affects plant growth. The main functions of mulch film are preventing weed growth, reduce the light transmittance, and prevent moisture loss of soil. Moreover, it is a barrier for soil surface also prevents soil compaction caused by rain which reduced the root damage. Plastic mulch is usually used for growing crops that have short harvesting time, such as cucumbers, strawberries, watermelons, etc. After that, the mulch film must be taken off to avoid plant diseases. (8)

Nowadays, the market of mulch film has promising increase of requirements. In 2020, the value of mulch film has more USD 4.07 billion which growing up to 6.5% from 2015. This tendency is the opportunities of increasing revenue and driving the market. (9) Moreover, agriculture market has growing the market size more than 10 billion in 2050. (1) However, the expansion of the market of mulch film causing the rapidly increasing of agricultural plastic waste. In the commercial, the mulch film made from synthetic material such as polyethylene (PE), low density polyethylene (LDPE). These synthetic mulch films cannot biodegradable also can combine in the soil for a long time and block of growth the plant root. (9) The mulch film is a single-use plastic and cannot recycle. The mulch film was used directly contact with the soil

therefore, the plastic has dirty and contaminate with other substance. Recycling of the mulch film is possible if the mulch film must contaminate less than 5%. However, in the field, mulch film was commonly contaminated from pesticide, herbicide, soil, and UV additive higher than 5% which up to 40-50% contaminate. (10)

For the long term uses, the synthetic plastic mulch film will get to the non-degradable plastic waste after the harvesting the crop. The residual mulch film can combine and remain in soil and ground water and thereby affect the environment. (2, 9) In the long term, these plastics have cracking and fragmentation to a small size which generates the microplastic in the environment. These impacts can lead to the reduction of soil and crop qualities. (5)

### **1. The mulch film in commercial**

The company and researcher have been interested to produce mulch film with the different color. They studied and investigated the appropriated color of the mulch film to increase the quality of the agricultural product.

#### **1.1 Black mulch film**

The black color of mulch film is widely use in climate zones. Black mulch film made from the synthesis material as low-linear density polyethylene with black pigment. (9) Black color can highly absorb the thermal energy and long-wavelength infrared. The use of the black mulch film affects to the increasing of soil temperature up to 6 °C and more moisture in the soil. (5) The black mulch film has a good efficiency in vegetable production due to the vegetable extends growing in the warm soil temperature. (2) Moreover, using the black mulch film cover on the soil could result in reduce of weed competition. (11)



**Figure 1 The black mulch film (12)**

### 1.2 Clear mulch film

The clear mulch film or transparent plastic mulch film has a UV transmittance of more than 85-95%. The clear mulch film has poor UV absorption which causing the soil has increase temperature from 4.4 to 7.8 °C. The UV-light was transmitted through the clear film and directly to the soil that affect the soil has less moisture in the soil. (13) The moisture in the soil has condensed and trapped in the film surface. The droplet was reflected the short-wavelength radiation and keep the heat in the tunnel of mulch film. This behavior is called the green-house effect. The clear mulch film has high transparency of radiation causing poor control of weed growth in the crop. (2, 5)



**Figure 2 The clear mulch film (14)**

### 1.3 White on black and silver reflective mulch film

Most of the white on black and silver reflective mulch film is used less than the black mulch film. This mulch film has similar properties to the black mulch film. The advantage of this mulch film can control weeds and appropriate to use in vegetable production. On the black mulch film can increase the soil temperature and weed management. On the white or silver can reflect the radiation thus can help the soil cooling. So, the white and silver on black mulch film have improved the soil temperature to suitable for the crop. (2, 13) Moreover, some literatures reported the silver on black mulch film influenced the insect repellent causing reduction the insect predation on vegetable production. (13)



**Figure 3 The white on black and silver reflective mulch film (15)**

The selection of mulch film for agricultural crops should be selected the suitable for the crop. The effect of the types of mulch film on the crops was summarized in Table 1. However, the mulch film has many benefits for crops and agriculturists. Although, these mulch films cause affect the environment. All synthesis mulch films are photodegradable materials that non-biodegradable. Synthesis mulch film was broken down after exposure to sunlight. The petroleum polymer was cracking causing release the of toxic chemicals such as phthalate, UV stabilizer, inorganic pigment. (5) The fragmentation of synthesis mulch film non-degradable causing generated many of microplastic and contaminated in the soil and groundwater. (13)



**Table 1 The effect of mulch film on the agricultural crops (11)**

<b>Crops</b>	<b>Types of mulch film</b>	<b>Crop effect</b>	<b>Environment effect</b>
Pepper	Black	Lower flowering	Increase soil temperature
	Silver on black	Reduce diseases	Reflect the UV radiation
	Clear	Increase yield	Increase soil temperature
Basil	Black	Increase yield	Decrease weed growth
Watermelon	Black	Increase growth rate	Increase soil temperature
Tomato	Clear	Increase yield	-
	Black	Early yield	Control soil temperature
Strawberry	Black	Increase yield	Elevated soil temperature
	Clear	Early yield	Increase soil temperature
Coffee	White	-	Decrease soil temperature
	Black	-	Increase soil temperature

## **Manufactory of mulch film**

Mulch film production has developed by using polyethylene (PE) since 1938. The mulch film comes to the commercial in 1939 which has the idea to produce for vegetable crops. (16) Manufacturers of mulch film can be processed into 2 designs including flat film and blow film. The flat film was processed by the die extrusion. Processing of flat film has quick processing and easier to control the thickness. However, the mulch film processing by the flat film has the limitation of the width of the film. Moreover, the condition has not stable to the process may cause the die extrusion to have a die swell. The blow film extrusion comes to solve these problems. In the general, the blow film extruder is widely processing in the manufactory of mulch film. Because the blow film extruder can generate the width film more than the flat film by die extrusion. (17) However, the selection of the mulch film processing depends on the customer requirement. This research has been interested to produce the mulch film by blow film extruder, which can separate into 2 steps including preparation of the compound by a twin-screw extruder. Then, lead the compound to produce the film by the blow film extruder.

### **1. Extrusion**

The word of “extruder” comes via New Latin from the Greek also “ex” meaning out and “truder” meaning push out which is the type of output called an extruder. (17) Extrusion is one technique for plastic processing. This technique pushes and develops the molten polymer through the die by pressure. Pressure in the extrusion is from the resistance between the rotation of screw with the barrel. The extrusion is the continuous process. The characteristic of die shape, flow properties of the polymer, and flow rate influenced the pressure to push the molten polymer through the die. (18) The production from extrusion covers various plastic products such as film, sheet, profile, pipe, wire coating, and other products. (17) Which, the machine of the extrusion is called an extruder. (18)

#### **1.1 Extruder**

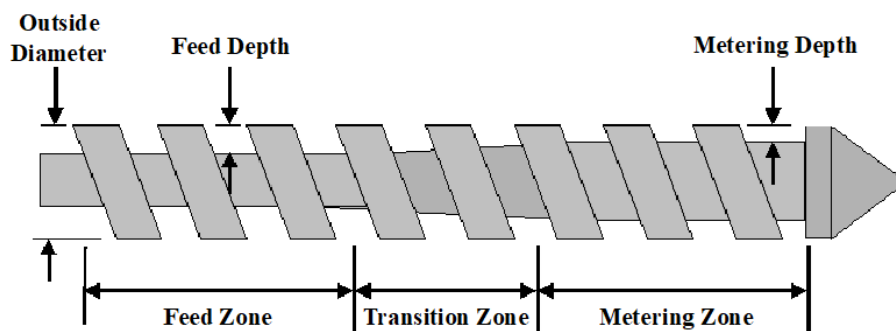
The extruder is the machine using in plastic processing with extrusion. The extruder was compounded the polymer materials and extruded the molten polymer through die that is the first part of the mulch film manufactory. Then, the compound was led to film production by the blow film extrusion. The extruder

consists of 3 zones in the barrel are feed zone, transition zone, and metering zone, respectively. (17, 18)

The first zone is the feed zone. This zone was carried the raw materials into the barrel. The screw has the deepest channel. The temperature of the feed zone has the lowest temperature. In this zone was carried the raw material and preheating. After that, the raw materials were compounding together. In this zone, the polymer has started to the molten polymer. (17-19)

The transition zone is the second zone of the screw. This zone has decreased the channel between screw and barrel. The diameter of the screw has increased from a small diameter in the feed zone. The polymer was to melt by compression with increasing shear rate from screw and barrel. The polymer melts were pushed to the metering zone for preparation to extrude through the die. (17-19)

The last zone is the metering zone. The deep channel and diameter of the screw have constant and less deep than every zone. In this zone was increasing the pressure causing push the molten polymer through the die. (17-19) Which can see the screw in Figure 4.

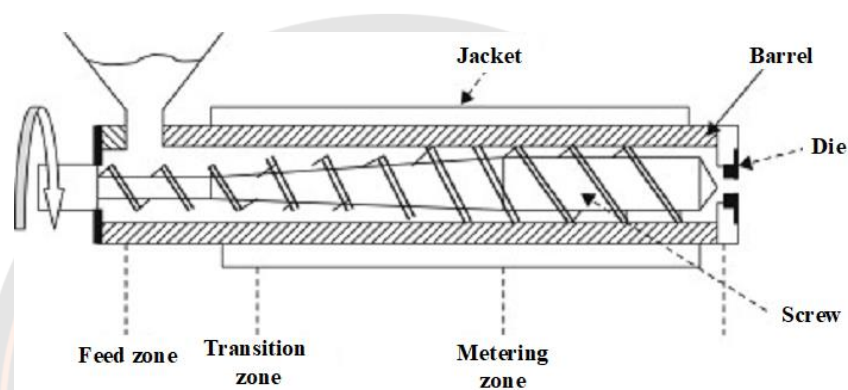


**Figure 4 The extruder screw zone (20)**

The extruder can be separated the type of extruder into 2 types are single screw extruder and twin-screw extruder. (18) The details can be illustrated as following.

### 1.1.1 Single screw extruder.

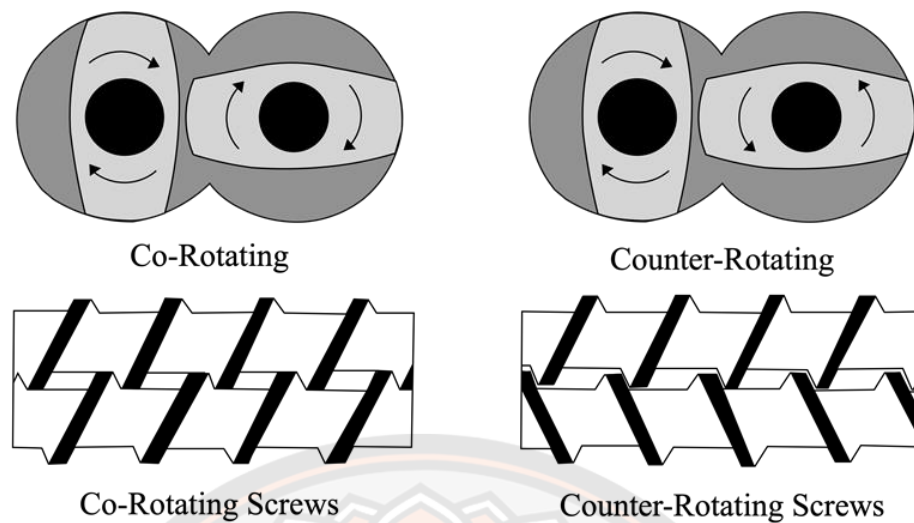
The barrel has contained just a single screw that rotating in the barrel. A single screw extruder has appropriate for simple production. The rotating of screw causing create the pressure to push the molten polymer through the die. Single screw extruder is most popular to produce the simple design and ruggedness. However, the mixing efficiency by a single screw extruder has poorly than a twin-screw extruder. The single screw extruder can see in Figure 5. (18, 21)



**Figure 5 Single screw extruder (20)**

### 1.1.2 Twin-screw extruder.

Twin-screw extrusion is a technique for mixing and continuous compounding polymer. Twin-screw extruder is widely used in the plastic manufactory. Because a twin-screw extruder has more efficiency to provide the homogeneous compound more than a single screw extruder. (21) The barrel contains a twin-screw that rotating in the barrel. It can separate the intermesh level of the screw with non-intermeshing and intermeshing. The rotation of the screw in the barrel can be classified into 2 types include co-rotating and counter-rotating. (19) The rotating of a twin-screw extruder can see in Figure 6. The advantage of each rotation of the screw was summarized in Table 2.



**Figure 6 The rotation of twin-screw extruder (22)**

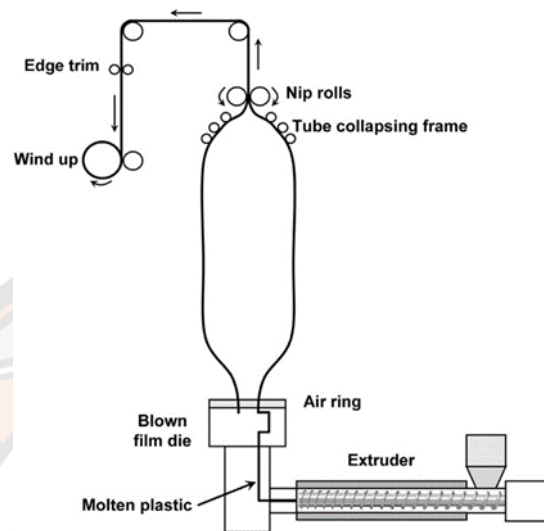
**Table 2 The summary of twin-screw rotations (23)**

<b>Co-rotating</b>	<b>Counter-rotating</b>
Rotating in same direction	Rotating in opposite direction
High speed of screw speed (up to 1800 rpm)	Limitation on the screw speed (~60 rpm)
High performance for compounding	Low performance for compounding
High melting efficiency	Low melting efficiency
Low of shear and heat	High of shear and heat

## 2. Blow film extrusion

Blow film extrusion is the second step after compounding the raw materials by a twin-screw extruder. Blow film extrusion is one technique for produce the plastic film. Blow film extruder is widely used to produce the plastic film. Because the blow film extrusion can generate the various diameter and faster production. (17) The processing start from the molten polymer was extruded through a tubular die. Which the tubular die is constant at  $90^\circ$  in the vertical direction. Then make the bubble film and cooling process with air from the cooling ring. The controlling of film thickness and diameter of bubble film can contribute by air pressure and nip process. The

extrusion of bubble film was pulled to the overhead of the blow film extrusion and after that was nipped by nip roll. The thin film was to keep on the winder. (17) The blow film production was seen in Figure 7.



**Figure 7 The blow film extrusion (24)**

## **Polymer blend and composite**

Hybrid materials have been interested in modern technologies. These materials have the properties of the hybrid with notable properties of each material. Polymer blend and polymer composite are one material that uses hybrid materials production. (25) The details of each technology were illustrated as following.

### **1. Polymer blend**

Polymer blend is one material for developing the technology of hybrid materials. The polymer blend can apply in widely application causing growth in various industries. Requirement of the market worldwide has increased at a rate of around 9% per year. The automotive industry, agricultural, aerospace, and packaging are the main market for polymer blend. (26)

The purpose of a polymer blend is to produce new materials by combining the efficiency of two or more components into one material. Properties of new material have hybrid properties corresponding to superior properties of each component. The compound after blending is called polymer blend. The polymer blend

has a single-phase that is homogeneous causing miscible blends. However, most polymer blends have an immiscible blend that leading to multiple phases in the morphology. (25, 26)

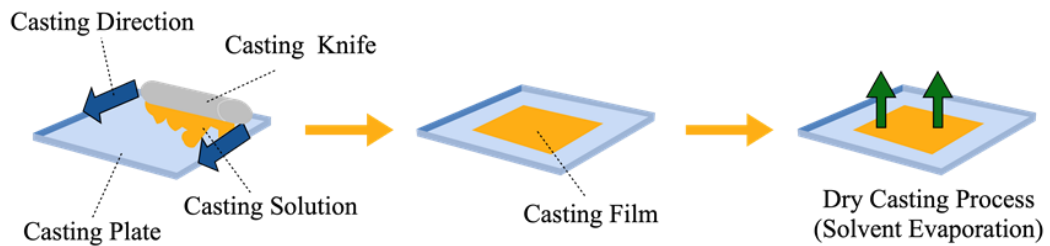
Miscible blends are a homogeneous phase that depends on the chemical structure, the molecular weight distribution, and the molecular arrangement. The blending between two-component has stable or metastable that depends on the suitable thermodynamic condition. Gibbs energy of mixing has a positive value causing expressed the stable blending. Moreover, investigate the miscible blends via the thermal properties that exhibit the single glass transition temperature ( $T_g$ ). (27) While the immiscible blend is a heterogeneous phase in the morphology. The multiple phases influence the interaction between the polymer that low adhesion when compared with a single phase. (26, 27)

#### 1.1 Blending method (26)

The blending method has a several different methods for blending the components of polymers to new materials. Selection method for blending that depends on the final properties that can perform into 2 processing include solution casting and melt blending.

##### 1.1.1 Solution casting

The solution casting is one method for film production. This method is mixing the solution of polymer and dissolved in a suitable solvent. Solvent were evaporated and driven from the polymer blend. The condition for processing that affect the film properties. The rate of solvent evaporation affects the mechanical properties and morphology of the polymer blend film. The solution casting process can see in Figure 8. However, the solution casting process is only suitable for laboratory scale. When up-scale for industry may caused the problem with the handling and control a large content of solvent.

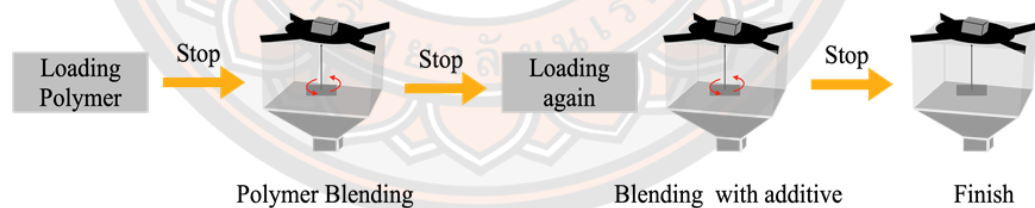


**Figure 8 Solution casting process**

### 1.1.2 Melt blending

Melt blending is widely used for the polymer blend process. Because this process is easier to up-scale for the industry. The melt blending with compounding 2 types are batch and continuous processes.

The batch blending using by the internal mixer. This process is necessary to the limited content of materials. The batch blending is appropriate for laboratory scale and a large scale. However, the disadvantage of batch blending is rather difficult to control the temperature in the chamber. In addition, batch blending has a processing step more than a continuous process. The batch blending process can see in Figure 9.

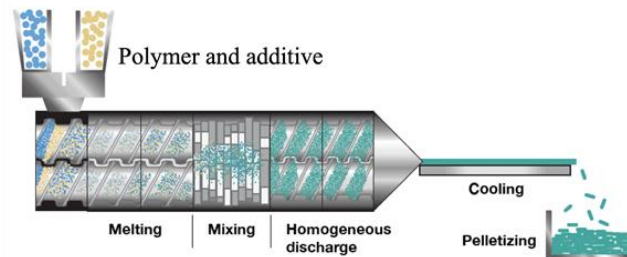


**Figure 9 Batch blending process**

The continuous process for polymer blends is easier to control production. The continuous process using the extruder that has various types of screw includes a single screw extruder and a twin-screw extruder. This process can up to a large volume of materials that easier to up-scale. This process can control the condition such as feed rate, screw speed, and temperature. The molten polymer of this



process has a homogenization. Last the molten polymer was extruded through the die. The continuous process can see in Figure 10.



**Figure 10 Continuous blending process (28)**

## 2. Polymer composite

Polymer composite composed of two or more materials and deform to the new material to improve the properties. The material used in polymer composite must be non-dissolved together. The physical properties of each material must be different. Polymer composite consists of two mains are matrix phase and reinforcement phase. (29)

The matrix phases are the continuous phase which has the volume more than the reinforcement phase. The matrix phases can classify into ceramic, metal, and polymer. (29) The selection of polymer matrix depends on the required properties. Most of the matrix phases are select from easy processing, good properties, and moderate cost. The strongly polymer composite are dependents on the properties of the matrix and reinforcement. (30)

The reinforcement phase is the discontinuous phase. The reinforcement can improve the mechanical properties of the polymer composite. The reinforcement material is usually strong and hard than the polymer matrix. However, the attribute of reinforcement also affects the properties of the polymer composite. (30)

The composite material is widely used in various industries such as aerospace, transportation, sports equipment, biomedical, and other applications.

## 2.1 Classification of polymer composite (30)

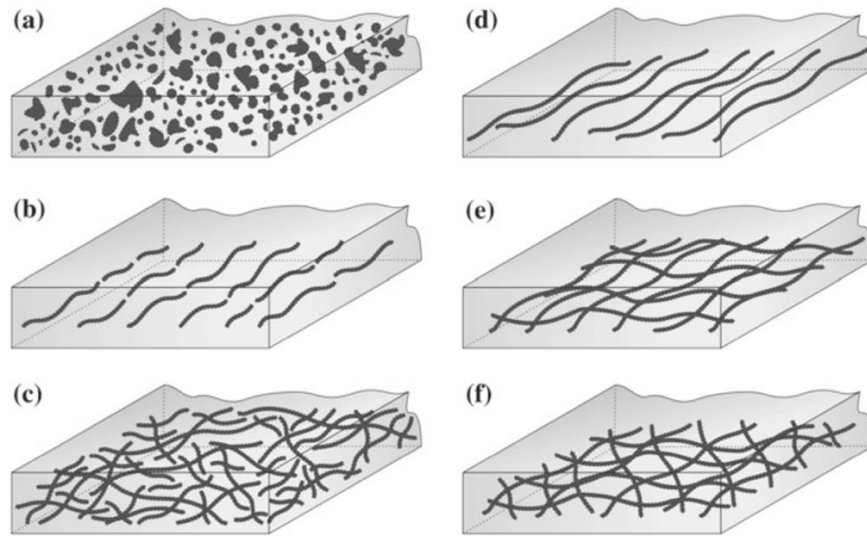
The polymer composite can classify follow the types of reinforcement materials. That can separate into 2 types consists of particle reinforces composite and fibrous reinforces composite.

### 2.1.1 Particle reinforces composite

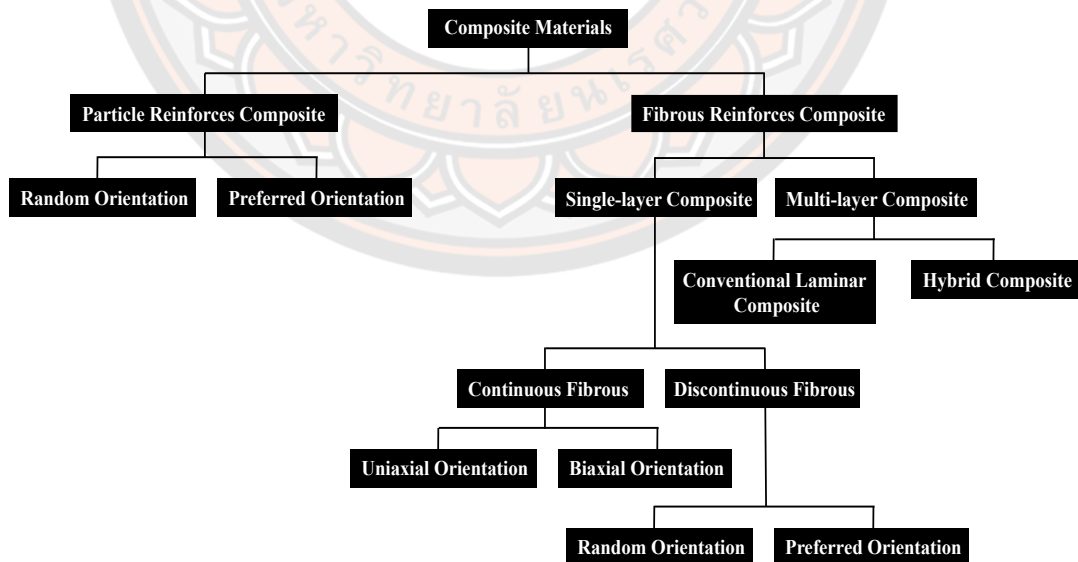
The particle reinforces material is the material with similar size that most particle has a small size. The particle shapes have circular, flake, and, cubic. Most particle reinforcements bind in the polymer matrix in random orientation. The arrangement of particles in the polymer matrix can see in Figure 11.

### 2.1.2 Fibrous reinforces composite

The fibrous reinforce has a length of more than the diameter. The diameter of fibrous was varied from 0.0025 mm to 0.13 mm that depends on the type of material. Which the fibrous can separate into continuous fibrous and discontinuous fibrous. The attribute of fibrous affects the orientation in the composite material and mechanical properties of the composite material. The arrangement of fibrous in the polymer matrix can see in Figure 11. That can be observed the classification in Figure 12.



**Figure 11** The fibrous orientation in the polymer matrix (a) Random orientation of particle, (b) Preferred orientation of discontinuous fibrous, (c) Random orientation of discontinuous fibrous, (d) Uniaxial orientation of continuous fibrous, (e) biaxial orientation of continuous fibrous, and (f) multi-direction orientation of continuous fibrous (30)



**Figure 12** Classification of the composite materials (29)

## **Bioplastic**

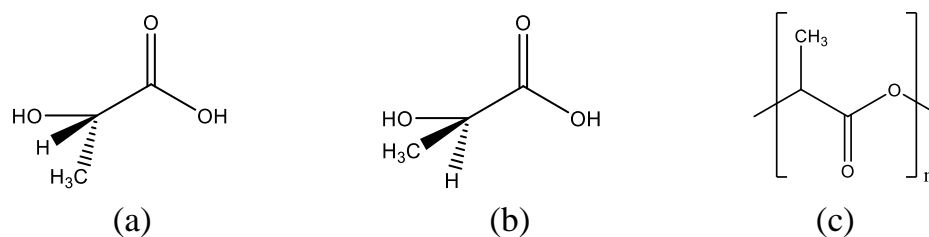
Bioplastic has been in use since the 1980s. Bioplastic is one type of polymer made from renewable resources. Bioplastic has been interested in various industries such as food packaging, agricultural, and medical. Bioplastic is one choice in production to replace petroleum plastic. Because petroleum plastic cannot decompose that generates a large quantity of plastic waste. While bioplastic can be degradable that through hydrolysis, photodegradation, fermentation, enzyme, and microorganism break down the chemical bonds in the polymer chain. (7, 31)

The widely use of bioplastic is aliphatic polyester. Biodegradation of polyester can be decomposed via hydrolysis. (7) In this work focus on the bioplastic 2 types include poly(lactic acid) and Polybutylene succinate.

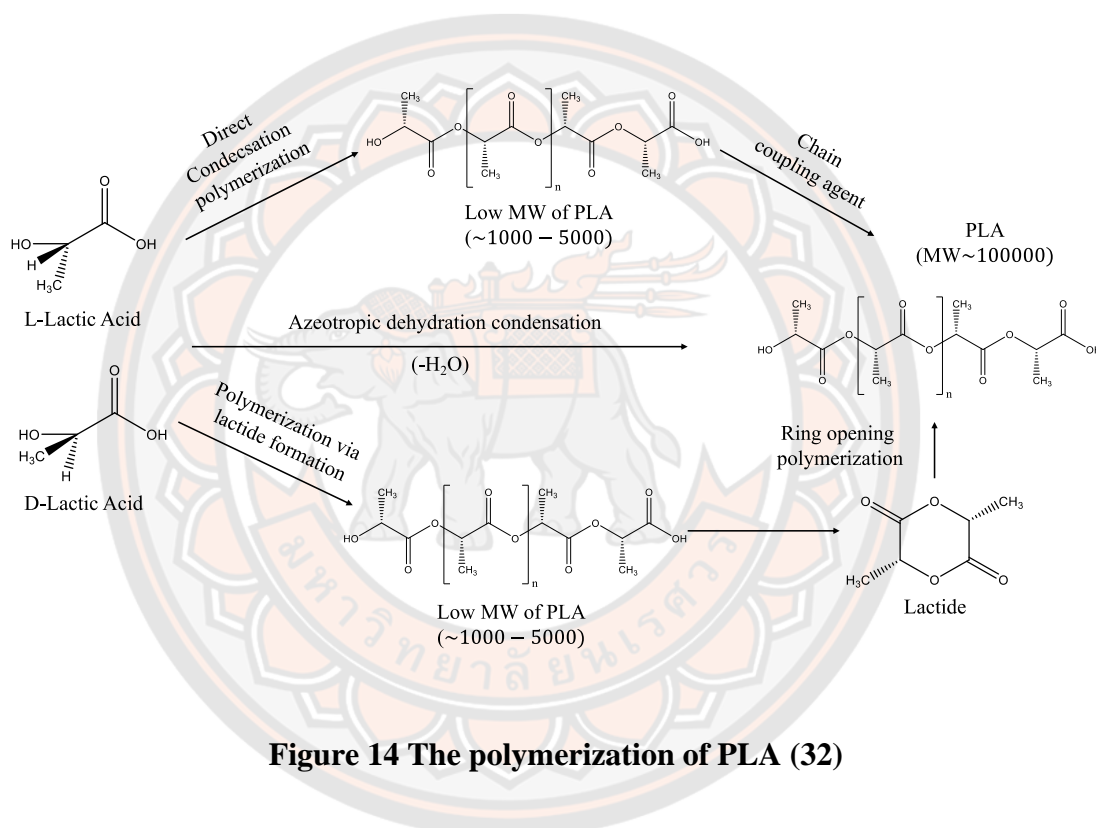
### **1. Poly(lactic acid)**

Poly(lactic acid) or PLA is a thermoplastic aliphatic polyester group made from renewable resources such as corn, cassava, and sugarcane, etc. Poly(lactic acid) is a semi-crystalline polymer that has crystallinity 37% therefore, PLA has transparency but it was hard and brittle. PLA has good thermal stability that has a high melting temperature which, is the glass transition temperature ( $T_g$ ) is 60-65 °c, and melting temperature ( $T_m$ ) is 170-180 °c. Moreover, PLA could be biodegradable and non-toxic so PLA has widely applied to use many applications for example, packaging, agriculture, and medical. (7)

Poly(lactic acid) can be divided into 3 types, that could be divided from different stereochemistry include Poly(L-lactic acid); PLLA, Poly(D-lactic acid); PDLA and Poly(D, L-lactic acid); PDLLA. The stereochemistry of PLLA, PDLA, and PLA monomer can be seen in Figure 13. The monomer of PLA is the lactic acid that, synthesis from fermentation of the corn, cassava, and sugarcane which, is the lactic acid was obtained from the fermentation process after that, the lactic acid was polymerized by ring-opening polymerization and another process to produce PLA could be polymerized by condensation polymerization which, have 2 types for the process include azeotropic dehydration condensation and direct polycondensation polymerization. (7) The polymerization of PLA can be seen in Figure 14.



**Figure 13 Stereochemistry of (a) PLLA, (b) PDLA, and (c) monomer of PLA (7)**



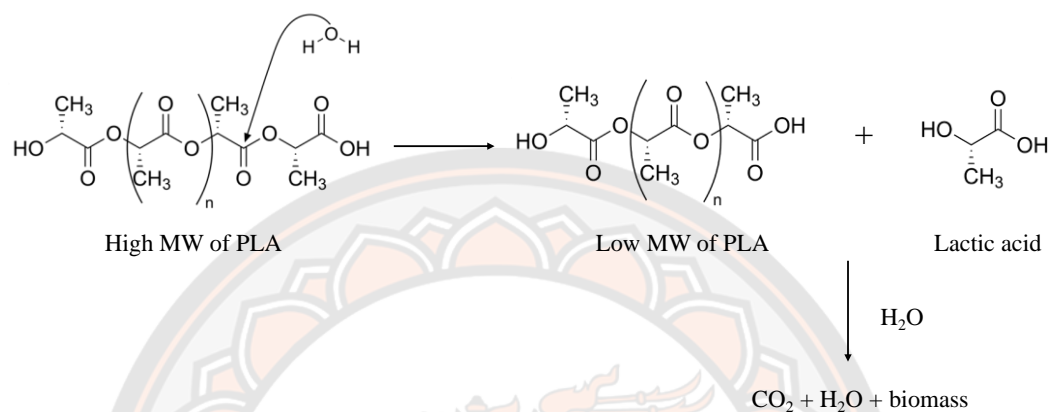
**Figure 14 The polymerization of PLA (32)**

### 1.1 Degradation of Poly(lactic acid) (33)

Poly(lactic acid) can be simply degraded by the hydrolysis of the ester bond. The degradation rate of PLA depends on many factors such as temperature, pH in the soil, humidity, quantity of oxygen, and microorganism. PLA after decomposing was reduced to CO<sub>2</sub> and water that non-releases the toxic chemical into the environment. In addition, PLA can decompose in different mechanisms apart from hydrolysis. These consist of photodegradation, and microbial degradation.

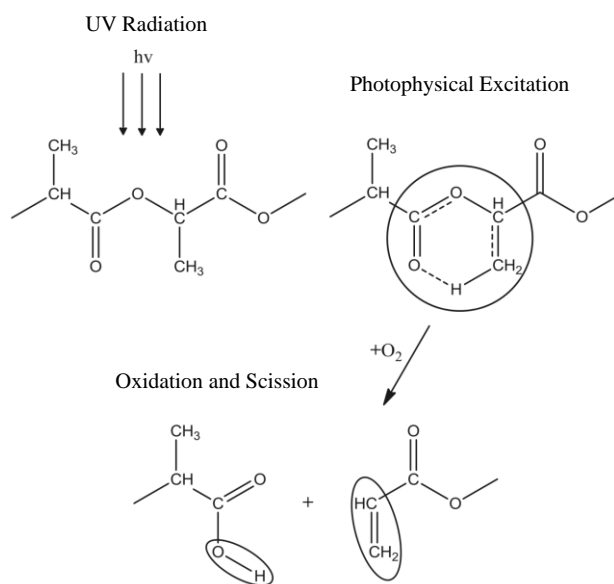
Hydrolysis degradation is the simple degradable of PLA. The ester group in the polymer chain of PLA is fragmentation and a decrease of molecular

weight. The polymer chain of PLA reacts with moisture causing the polymer chain was cleavage. The cleavage reaction was decreased in amorphous regions and increase in crystallinity. The hydrolysis degradation mechanism can observe in Figure 15.



**Figure 15 Hydrolysis degradation mechanism of PLA (34)**

Photodegradation is the degradable of PLA by UV radiation. The main chain of PLA was absorbed the UV radiation causing the backbone of the PLA chain to have a photochemical reaction. The UV radiation reacts within the PLA polymer chain and caused the chain scission by the Norrish reaction. The photodegradation mechanism can observe in Figure 16.



**Figure 16 Photodegradation mechanism of PLA (35)**

Microbial degradation is using the microorganism for decomposing of PLA. The microbial degradation was initiated under hydrolysis degradation. The microorganism creates the extracellular for depolymerase to break down of PLA chain. Which the ester bond is the target for depolymerase. The last products by microbial degradation of PLA are CO<sub>2</sub>, water, or methane. However, the long-times necessary for degradation because PLA has the resistance to microbial.

### 1.2 Application of Poly(lactic acid)

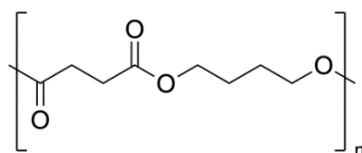
Nowadays, PLA was used in various industries such as packaging, medical, and agriculture. (7) The other work, PLA is on bioplastic that has been interested to study and improve the properties of PLA for applied to the suitable application.

N. Suaduang and co-workers (36) have been interested to produce the agricultural film from PLA incorporated with spent coffee grounds (SCG). To study the effect of SCG on the physical and mechanical properties of the bio-composite film. The film production using a twin-screw extruder and the blow film extruder. From the results were found the PLA and SCG have miscibility. The addition of SCG into the bio-composite film help to improve the flexibility of PLA. The bio-composite film can apply to produce a nursery bag.

Soo Hyun Sung and co-workers (37) have been interested to develop the nanocomposite film from the PLA with reinforcement cellulose nanocrystals (CNCs) from coffee silverskin (CS). To study the effect of CNCs on the mechanical and barrier properties of the nanocomposite film. The twin-screw extruder was used for nanocomposite film production. The results were found CNCs can improve the barrier properties and mechanical properties of the nanocomposite film. That less than 5% of CNC help to increase the tensile strength. The addition of CNCs less than 5% has the diffusion of water vapor and oxygen through nanocomposite film at a low rate. In this work, the nanocomposite film was developed and applied to use in packaging material.

## 2. Polybutylene succinate

Polybutylene succinate or PBS is an aliphatic polyester same PLA that, the chemical structure consists of a monomer is butylene succinate ( $C_8H_{12}O_4$ ) which, PBS processability synthesis via polymerization of succinic acid and 1,4- butanediol. The chemical structure monomer of PBS can observe in Figure 17. Polybutylene succinate was obtained from fossil-based resources and bio-base. The advantages of Polybutylene succinate could be biodegradable by fermentation and the mechanical properties are comparable to polyethylene (PE) that the thermal properties and tough higher more than PE. Polybutylene succinate is a non-transparency polymer. Polybutylene succinate has crystallinity 33-45 %, glass transition temperature ( $T_g$ ) is - 32 °c, and melting temperature ( $T_m$ ) is 114-115 °c. For the applications of PBS include food packaging, mulch film, plant pots, and medical, etc. (7)

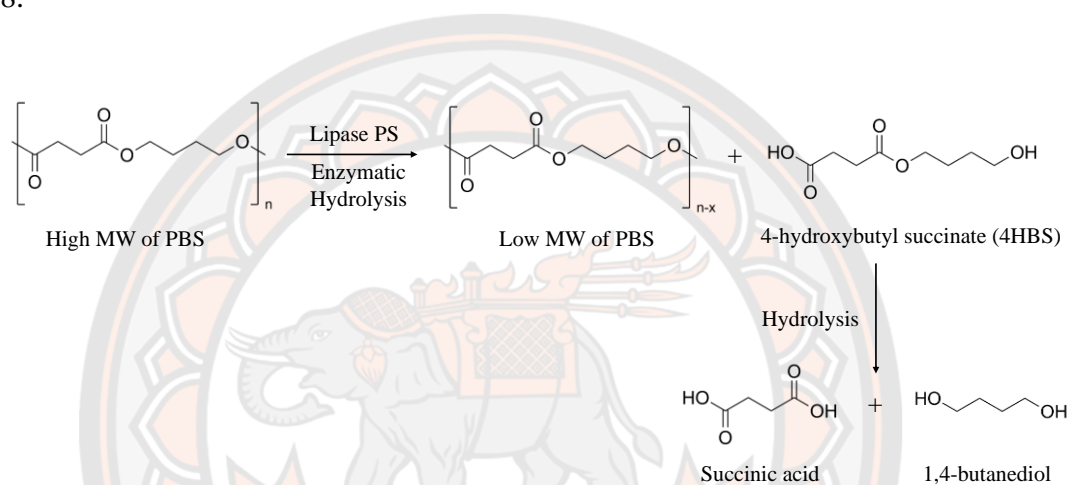


**Figure 17 Chemical structure monomer of PBS (7)**



### 2.1 Degradation of Polybutylene succinate (38)

The biodegradation of PBS through enzymatic degradation. The enzymatic degradation for PBS using *Pseudomonas cepacia* (lipase PS<sup>®</sup>). The lipase PS<sup>®</sup> act as the catalyze. After the PBS degradation by enzymatic degradation with lipase PS<sup>®</sup> gave the main product is 4-hydroxybutyl succinate (4HBS). Then 4HBS can be transfer to succinic acid and 1,4-butanediol by hydrolysis that reacts at ester bond of 4HBS. The enzymatic degradation mechanism of PBS can observe in Figure 18.



**Figure 18 Enzymatic degradation Mechanism of PBS (38)**

### 2.2 Application of Polybutylene succinate

Nowadays, PBS was used in various industries such as packaging, medical, and agriculture. Moreover, the more research has interested to improve the flexibility of the polymer blend by blending with the PBS. (7)

N. Hongsriphan and co-workers (39) have developed the composite material from PBS with lemon basil particle (LB). The addition of LB acts as moisture sorption. Moreover, loading ammonia sulfate into composite material as fertilizer. Moisture sorption and fertilizer releasing compared with the pure PBS film were investigated. The composite material produced using a twin-screw extruder for compounding and prepared composite film by hot pressing machine. The results were found the increasing LB content in the composite film can improve the moisture

sorption of the composite film and ammonia sulfate can faster release in water. Moreover, the addition of LB can help the degradation of PBS.

A. Bhatia and co-workers (40) have been interested to produce the blend film between PLA and PBS for advances in packaging material. Mechanical properties and compatibility of these polymers were examined. The polymer blend has production from a twin-screw extruder and made the blend film by compression molding. The results were summarized the polymer blend with PBS more than 20% causing the polymer blend film to have incompatibility. The polymer blend film with PBS 80% and 90% has properties like PBS that increases of %elongation. The polymer blend with PBS less than 20% can improve the deficiency of the PLA that improves the polymer blend film to flexible.

### **Natural fillers**

The natural filler is one classification of the additive. Natural fillers are covered base on plants, minerals, and animals. Plant filler has composed of cellulose as the main composition. Plant fillers are derived from renewable resources which can be extracted from leaf, bast, and wood from plants. The selection for applied rely on the required performant of filler. (30) The chemical composition of plant fillers composed of cellulose, hemicellulose, pectin, and lignin. These chemical compositions have hydroxyl components that distributed throughout the fiber wall of plants. However, the quantity of each component depends on the types of plants, sources, and processes of sample preparation. (41)

Natural fillers are particles add to plastic that can improve some properties of polymer which, are from nature that have many types such as particles, fibers, and nanofibers. The advantages of natural fillers are non-toxic, biodegradable, and non-remain in the product or environment. Moreover, some natural fillers exhibited antibacterial, antioxidant activities, therefore, the current scientists are interesting to used natural fillers instead of synthesis fillers, and the key point of using natural fillers will also usually reduce the cost of the base polymer.

The general, natural fillers must be stable through the processability and usability, non-toxic, and the main point is non-change the chemical structure of a polymer and it can disperse and compatibility with a polymer. The shape and size of

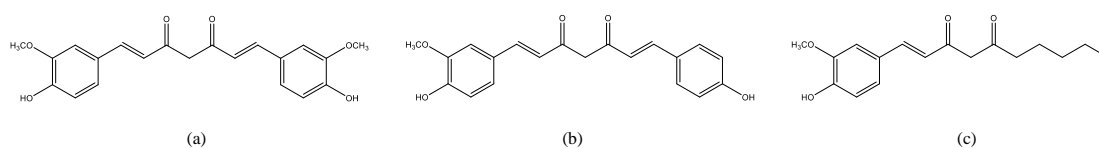
particle fillers are important. The smaller particles can be improving the value of properties such as tensile strength, modulus, and hardness. The larger particles can be giving less strength to a polymer.

### 1. Ginger

Ginger is one of the spices used as a flavoring agent. The original ginger from India and become to Africa and Caribbean. The different locations for cultivation also affect the quality of the ginger as flavor and appearance. Moreover, the harvesting time and maturity of plant also affect the quality of ginger such as the moisture, chemical compositions including non-volatile compounds, and volatile compounds. (42)

Ginger (*Zingiber officinale* Roscoe) belongs to the Zingiberaceae family. It is a monocotyledonous herbaceous perennial which grows annually in Southeast Asia. It is a native plant in Indonesia, India, Malaysia, and Thailand. (43) Ginger can be divided following harvesting period of crop include immature ginger (fresh ginger) has age 5 months, early immature ginger has age 5-7 months, and mature ginger has age 8-9 months. Ginger has a small scale of leaf and most of the scale is a branch of root that is rhizomes. The cross-section of rhizomes ginger shows the pale-yellow color. The intensity of color depends on the maturity stage of ginger. (42) The pigments of yellow color known as curcuminoids. The major pigments consist of curcumin, demethoxycurcumin and 6-dehydrogingerdione. The intensity of these compounds including depends on the maturation stage of ginger. (44)

Moreover, the non-volatile compound of the ginger consists of oleoresin as a flavor component that gingerol and shogaol are spicy component. The ginner also has the volatile compound as essential oil 1-3% which affects the sensation of smell and taste. Moreover, gingerols are major pungent constituents of ginger which are made up of several different compounds. Gingerols can be classified as shogaol, paradol, and zingerone. (44, 45) The chemical structures of active components of ginger can see in Figure 19.

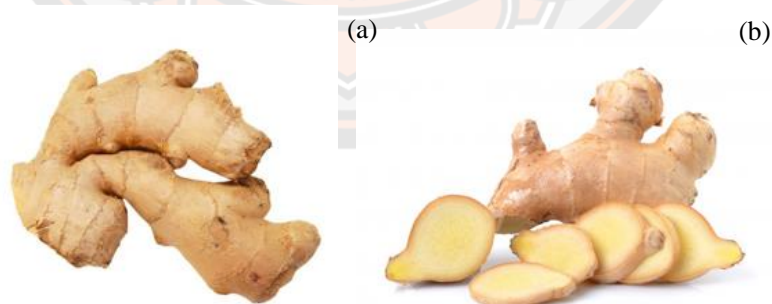


**Figure 19** The chemical structures of active components from ginger; (a) 6-gingerol, (b) 6-shogaol, and (c) 6-paradol (45)

This research has been interested to study the different maturation stages of ginger. The develop the ginger as a natural filler for applied in the bio-composite film. However, the chemical composition as fibrous, color shade, and moisture depend on the maturation of ginger. So, this work focused on the information of each maturation stage of ginger that separated into 2 ages are early immature ginger and mature ginger.

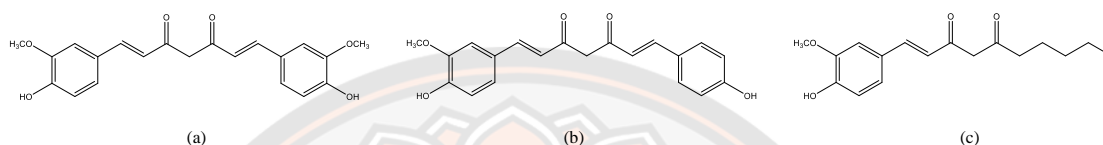
#### 1.1 Early immature ginger

The early immature ginger has harvest around 5-7 months after cultivation. The appearance of the peel ginger has obviously no withered, and the peel color appears the pale-yellow shade. The cross-section of ginger has pale-yellow and less fibrous that can observe in Figure 20. (44)



**Figure 20** The appearance of (a) peel ginger and (b) the cross-section of early immature ginger (46)

The pigment compounds of early immature ginger consist of curcumin 1.23 mg/100 g of ginger, demethoxycurcumin 0.77 mg/100 g of ginger, and 6-dehydrogingerdione 6.67 mg/100 g of ginger. Moreover, the investigation of the color parameter as  $b^*$  value appears approximately  $11.67 \pm 3.79$  according to the positive value that attributes a yellow shade. (44) The chemical structures of curcuminoids and hydroxycinnamic acid derivatives are shown in Figure 21.

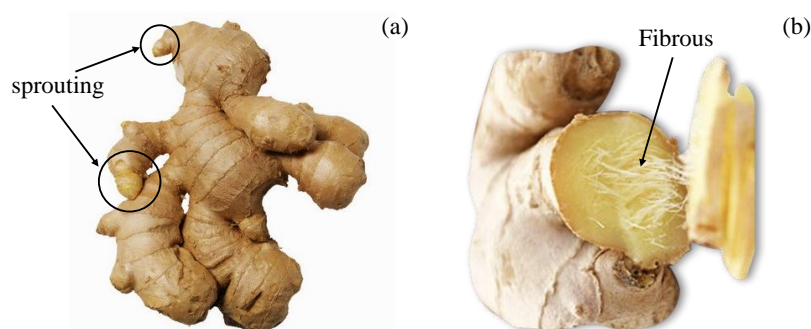


**Figure 21 The chemical structure of curcuminoid compound as (a) curcumin, (b) demethoxycurcumin, and (c) 6-dehydrogingerdione (44)**

Furthermore, the early immature ginger has more intensity of moisture than mature ginger. The other research has studied the quantity of moisture and crude fiber of early immature and mature ginger. It was found early immature ginger has the content of moisture as 91.61%. The crude fiber of early immature ginger has 1.35 g/100g of ginger. (47) Early immature ginger is most popular to use the preserved ginger because it also remains tender and mild. (42)

### 1.2 Mature ginger

The mature ginger has ages around 8-9 months after planting. The external appearance of mature ginger rhizome presents a more intensity of yellow shade than the early immature ginger. The inside rhizome of mature ginger has a more fibrous and yellow color. (44) Moreover, the mature ginger has more notice is started to be sprouting, which can observe in Figure 22. The appropriate of mature ginger should be harvesting that observes the leaf begin to turn yellow and become dry. If the harvest has delay causing the rhizome has increase sprouting. (42)



**Figure 22 The appearance of (a) peel ginger and (b) the cross-section of mature ginger (46)**

The color of mature ginger rhizome has more intensity than the early immature ginger thus causing the pigment to have the greater chemical compounds. The yellow pigments consist of curcumin increase to 2.47 mg/100 g of ginger, demethoxycurcumin high to 1.30 mg/100 g of ginger, and 6-dehydrogingerdione 12 mg/100 g of ginger. The increasing of the quantity of these pigments causing the intensity of yellow was also increased. The  $b^*$  value of the mature ginger has  $91 \pm 4.51$  that higher than the early immature ginger. (44)

The mature ginger rhizome has a content of fiber higher than the early immature ginger. The report presents the mature ginger has crude fiber 1.52 g/100 g of ginger. On the other hand, the moisture in mature ginger was reduced to 88.80%. (47) The quantity of the chemical compounds is non-volatile, and the volatile compound does not find the report. However, probably these compounds promising an increase following the maturation stage of ginger. Mature ginger has popularly used as ginger powder for spices and medicines. Moreover, the essential oil in mature ginger has been interested to extract and apply in medical field. (42) The comparison of compositions of the different maturation stages of ginger were summarized and shown in Table 3.

**Table 3 Compositions of different maturation stages of ginger**

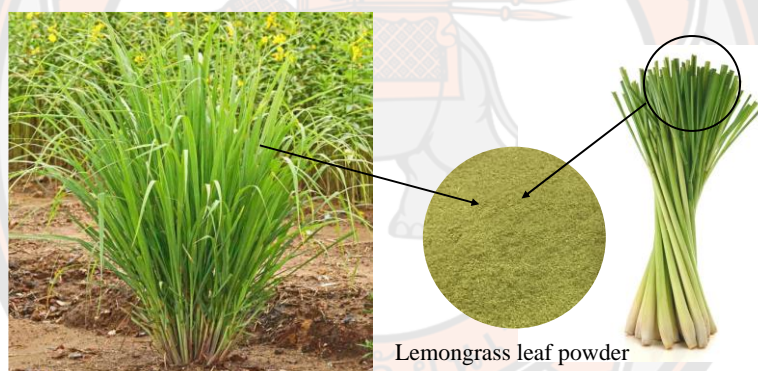
Composition	Early immature ginger	Mature ginger	Ref.
Age	5-7 months	8-9 months	(42)
Crude fiber	1.4% at least	up to 9.5%	(42)
Total ash	6.11% at least	up to 9.58%	(42)
Crude protein	8.1% at least	up to 11.6%	(42)
Starch	41.54% at least	up to 55.06%	(42)
curcumin	1.23 mg/100g	2.47 mg/100g	(44)
demethoxycurcumin	0.77 mg/100g	1.30 mg/100g	(44)
6-dehydrogingerdione	6.67 mg/100g	12 mg/100g	(44)
b* value	11.67 ± 3.79	91 ± 4.51	(44)

The ginger can apply in various application. Mostly use of ginger is spices that give the flavoring test and good smell. Moreover, the ginger applied in the medical uses and cosmetics due to ginger has many active compounds for anti-inflammation and antimicrobial activity. (45) This research focused on the fiber form ginger that applied in the composite material.

J. Jacob and co-workers (48) have been produced and developed the nanocomposite with cellulose nanofibers from ginger rhizomes (GNF). They prepared the nanocomposite film by solvent cast method. Polyvinyl chloride (PVA) and chitosan (CS) were used for the polymer matrix. The aim was to study the effect of GNF on the mechanical properties and antimicrobial for applied to the biomedical application. The results were found the addition of GNF at 5% to CS and PVA matrix help improved mechanical properties. While increasing of the tensile strength of the nanocomposite film because GNF has a good interaction with PVA and CS matrix by a hydrogen bond. Moreover, GNF has a bacterial inhibition ability that can inhibit in the positive gram and negative gram.

### 3. Lemongrass

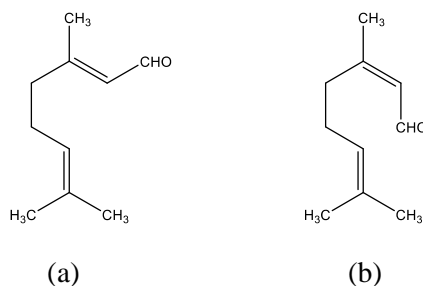
Lemongrass is an herbaceous perennial that grows into clumps up to high 3 m that can see the attribute of lemongrass in Figure 23. The scientific name is *Cymbopogon citratus* (DC.) Stapf which is belonging to Gramineae (Poaceae) family. Lemongrass grows annually in Sri Lanka and India. (49) Lemongrass consists of water approximate 80%, essential oil 0.2- 0.4% and it may be found to higher 3% depended on extraction method and source of material. Moreover, it has another compositions and minerals such as calcium and potassium. (50) The harvest time of lemongrass at intervals of 60-70 days after planting. (51) Lemongrass was used for various applications. This plant was widely used as flavoring agent in food for cooking. (49) Moreover, lemongrass was used as the source for extraction of essential oil and was produced as lemongrass powder.



**Figure 23 Lemongrass (*Cymbopogon citratus*)**

The main chemical component of lemongrass is citral which has two isomers consists of citral a (geranial), and citral b (neral) (Figure 24). These compounds are abundant found in essential oil. In addition, the processing to make of lemongrass powder may cause the reduction of citral content due to its thermal-sensitive property with high temperature. The report exposes that the extraction of citral compound from lemongrass powder with approximately 150 mg/g. (49) Moreover, the lemongrass powder also contained the other group of phytochemical compounds such as phenolic acids and flavonoids. (52)





**Figure 24 Chemical compound of lemongrass (a) citral a (geranial) and (b) citral b (neral) (53)**

This research focused on the lemongrass leaf due to the lemongrass leaf is the agricultural waste after harvesting and not yet take benefit. Moreover, lemongrass also contained cellulose, hemicellulose, and lignin as the compositions. Their content was reported as cellulose 39.5%, hemicellulose 22.6%, and lignin 28.6%. (54) Lemongrass contained chlorophyll A and chlorophyll B as the pigments and responsible for its green color. In the study of Thorat PP and co-worker, they found a total content of chlorophyll equal 0.1338 mg/g of lemongrass powder. The color parameter has  $b^*$  value as 27.28 were reported. (55)

Lemongrass is mostly used as flavoring ingredient for food. Moreover, lemongrass was taken in the cosmetics, pharmacy, and medical because it has a more active compound for antioxidant and antimicrobial activity. (51) This research has been interested to use the lemongrass for development of the composite materials.

From the literature, L. D. Bekele and co-workers (54) have been studied and developed the thermoplastic composites reinforcement with lemongrass powder. They prepared the composite from high-density polyethylene (HDPE) and lemongrass powder compounding by melt blending method with a twin-screw extruder. To study the effect of lemongrass powder as reinforcement on the mechanical properties and thermal properties, the results show the addition of lemongrass powder at 10% can improve the tensile strength. In addition, lemongrass powder help to improve the flexural properties of thermoplastic composites. The study of thermal property of thermoplastic composites found that the degradation temperature shifts to a higher temperature when adding of lemongrass powder. In the summary, lemongrass powder

can improve the mechanical properties in term of reinforcement, and it can improve the stability of temperature degradation.



## CHAPTER III

### RESEARCH METHODOLOGY

The mulch film manufactory was performed in 2 steps. The first step was to produce the bio-composite compound by using a twin-screw extruder. Then lead the bio-composite compound was to perform the bio-composite film via the blow film extruder. To purpose the effect of the natural fillers on the chemical properties, physical properties, mechanical properties, and biodegradability. The details are as following.

#### Raw materials

1. Ginger rhizomes from the local market in Phitsanulok, Thailand.
2. Lemongrass leaf from the local market in Phitsanulok, Thailand.
3. Poly(lactic acid); PLA with Ingeo™ Bio-polymer 4043D grade from NatureWorks LLC.
4. Polybutylene succinate; PBS with BioPBS™ FD92PM grade from NatureWorks LLC.

#### Equipment

1. Universal oven, Memmert Model UN110 from BEC THAI, Thailand.
2. Precision scale balance 2 points, DENVER INSTRUMENT Model TP-2102 from BEC THAI, Thailand.
3. Precision scale balance 4 points, DENVER INSTRUMENT Model SI-234 from BEC THAI, Thailand.
4. Laboratory test sieve 325 mesh (45 micron), Endecotts according to ISO 85 EN 9001.
5. Glass bottle with diameter 4.0 cm.
6. Grinder 1500 W, COSUAI Model CS-700.
7. Desiccator.

## **Instrument**

1. Small scale twin-crew extruder, LABTECH Model LTE16-40.
2. Small scale blow film machine, LABTECH Model LE20-30/C & LF-250.
3. Fourier transform infrared spectrometer (FT-IR), Perkin Elmer Model Spectrum GX.
4. X-ray diffractometry (XRD), BRUKER Model D2 Phaser.
5. Scanning electron microscope (SEM), Model Leo1455VP.
6. Calorimeter, Konica Minota Model CR-20.
7. UV-vis spectrophotometer, Specord 210 plus.
8. Universal testing machine, INSTRON<sup>®</sup> CALIBRATION LAB Model 5965.
9. Differential scanning calorimeter (DSC), METTLER TOLEDO.
10. Melt Flow Index (MFI), Instron<sup>®</sup> CEAST Model MF20.

## **Methodology and characterization technique**

The research methodology and characterization in this work was separated into 4 sections. The detail as following.

### **1. Natural fillers**

#### **1.1 Preparation of the natural fillers**

Early immature ginger powder (EG), mature ginger powder (MG), and lemongrass leaf powder (LE) were cut to and small size of approximately 1-2 cm and removing the dirty by water. Let dry the pieces of EG, MG, and LE by the universal oven at 60 °c for 24 hr. Then, the dried EG, MG, and LE were reduced size by using the grinder. After that, let sieve by the laboratory test sieve with 45 microns. All the natural fillers were stored in a plastic seal bag and keep in the desiccator to prevent moisture.

#### **1.2 Characterization of the natural fillers**

The whole of natural fillers was characterized into 3 properties. The chemical properties, morphology, and Fiber length were studied. The detail can be seen as following.

### 1.2.1 Chemical properties

The natural filler was indicated the functional group by using Fourier Transform Infrared Spectroscopy (Perkin Elmer model Spectrum GX) with ATR mode. The whole of natural fillers were analyzed in the wavenumber region of  $4000 - 400 \text{ cm}^{-1}$ .

### 1.2.2 Morphology

The natural filler with particle size around 45 microns was analyzed the morphology by using a scanning electron microscope (Model Leo 1455VP, CARL ZEISS CO., LTD.). The each of natural fillers was put on the stub with carbon tape. After that, the sample was sputtered with gold to improve the conductivity of the sample surface. The evaluation was indicated at magnifications as 200X and 1000X, respectively.

### 1.2.3 Fiber length

The SEM image of natural filler was investigated fiber length of natural filler by ImageJ software with version of Fiji using Java 6. The measurement of fiber length was measured with 100 units of each natural filler.

## **2. The bio-composite film from poly(lactic acid) incorporated with natural fillers**

### 2.1 Preparation of the bio-composite film

The bio-composite film was performed into 2 steps. The first step was prepared the bio-composite compound. The second step was produced the bio-composite film.

#### 2.1.1 The bio-composite compound

The bio-composite compound was performed from poly(lactic acid); PLA incorporated with various content of the natural fillers by a twin-screw extruder. The raw materials were removed moisture by the universal oven at  $60 \text{ }^{\circ}\text{C}$  for 2 hr before compound process. The temperature condition for compounding was used 100, 120, 140, 150, 160, 170, 180, 180, 180, and  $170 \text{ }^{\circ}\text{C}$  with screw speed 100 rpm. The ratio of the bio-composite compound between PLA and natural fillers was assigned in Table 4.

### 2.1.2 The bio-composite film

The bio-composite compound pellets from a twin-screw extruder was produced the bio-composite film by the blow film extruder. The condition of blow film was set at the temperature as 150, 170, 180, and 170 °c and screw speed 90 rpm. The obtain bio-composite film has a thickness of 60-70 microns and a width of 17 cm.

**Table 4 The ratio of the bio-composite film between poly(lactic acid) and each natural fillers**

Bio-composite	Weight (g)	
	Poly(lactic acid)	Natural filler
Neat PLA	100	0
	97.5	2.5
	95.0	5.0
EG	92.5	7.5
	97.5	2.5
	95.0	5.0
MG	92.5	7.5
	97.5	2.5
	95.0	5.0
LE	92.5	7.5
	97.5	2.5
	95.0	5.0

## 2.2 Characterization of the bio-composite compound

The bio-composite compound was to assay into 2 properties. The characterization was studied into morphology and the melt flow index. The detail can see in as following.

### 2.2.1 Morphology

The bio-composite compound pellet was analyzed the cross-section morphology after cutting from a twin-screw extruder. Scanning electron microscope (Model Leo 1455VP, CARL ZEISS CO., LTD.) was used to

characterization. The pellet was stick on the stub with carbon tape and coating with gold. The morphology characterization with 200X and 1000X as magnification.

### 2.2.2 Melt flow index (MFI)

The bio-composite compound pellet from a twin-screw extruder was evaluated the melt flow index via melt flow indexer (Instron<sup>®</sup> CEAST MF20). The melt flow testing was according to ASTM D1238E. Preparation of the bio-composite compound pellets approximately 8 g with testing under temperature conditions as 190 °c and 2.16 kg as load.

### 2.3 Characterization of the bio-composite film

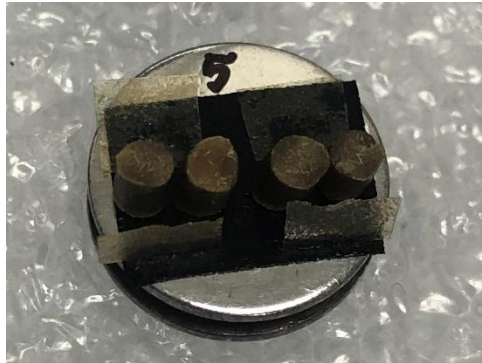
The bio-composite film was assayed into 4 properties. The characterization was studied on the chemical properties, physical properties, mechanical properties, and biodegradability. The detail can see as following.

#### 2.3.1 Chemical properties

Fourier Transform Infrared Spectroscopy (Perkin Elmer model Spectrum GX) was used for determining the functional group of the bio-composite film. The bio-composite film was assayed with ATR mode and scanning in the wavelength from 4000 – 400 cm<sup>-1</sup>.

#### 2.3.2 Physical properties

**Morphology:** The surface morphology of the bio-composite film was scanned by scanning electron microscope (Model Leo 1455VP, CARL ZEISS CO., LTD.). Cutting the bio-composite film around 3 x 3 mm. The film specimen was put on the stub by using carbon tape and gold sputtering on the sample. Moreover, investigate the cross-section morphology of the bio-composite film from tensile testing. The sample was put on the stub which the area cracking of the bio-composite film on the top view. The magnification was scanned at 200X and 1000X. The preparation of the bio-composite film can see in Figure 25.



**Figure 25 Preparation of the sample of the bio-composite compound and bio-composite film for SEM analysis**

Water vapor permeability (WVP): The bio-composite film was assayed the water vapor transmission (WVTR) following ASTM E96-80. The glass bottle with a diameter of 4 cm contains silica gel in the bottle. Then, lead the bio-composite film has covered on the glass bottle and sealed. The sample was weighed of the silica gel for the first time as a starts weight. The sample was placed in a desiccator at 25 °c and 65 % RH as humidity. The water vapor transmission rate was evaluated by the weight change of the silica gel in a glass bottle every 24 hr for 7 days. Which the weighing was determined with a precision of 0.0001 g. The water vapor transmittance rate (WVTR) was calculated as follows.

$$\text{Water vapor transmission (g/m}^2 \cdot \text{day)} = \frac{G/t}{A}$$

Where: G/t = Slope of the straight line between weight changes and time changes (g/day)

A = Test area which is mouth of glass bottle area (m<sup>2</sup>)

Color parameter: The bio-composite film was measured the color parameter by using a calorimeter (Konica Minolta model CR-20) following ASTM E313-96. The bio-composite film with an area of 15 x 15 cm was identified in 10 different positions. The data report was obtained 3 parameters consists of L\*



(lightness),  $a^*$  (red-green), and  $b^*$  (yellow-blue). Color difference ( $\Delta E$ ) can be expressed as the value of the sample compared with the sample standard. The calculation of  $\Delta E$  can be as follows.

$$\Delta E = \sqrt{(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2}$$

Where:  $\Delta L^*$  = Different of lightness and darkness (+ as lightness, - as darkness)

$\Delta a^*$  = Different of red shade and green shade (+ as red shade, - as green shade)

$\Delta b^*$  = Different of yellow shade and blue shade (+ as yellow shade, - as blue shade)

UV transmittance: The bio-composite film was measured %UV transmittance via UV-Vis spectrophotometer (specord 210 plus). The film specimen was cut to a size of 3 x 3 cm which prepared 3 pieces of the sample. That recorded in wavelength on 200-700 nm with scanning rate as 50 mm/min.

### 2.3.3 Mechanical properties

The bio-composite film was assayed the mechanical properties by Tensile testing according to ASTM D638 with dumbbell Type I. Using universal testing machine (INSTRON® CALIBRATION LAB model 5965). Preparation of the specimen as dumbbell form with 7 specimens. Testing under the condition with load cell 1 kN, tension rate 20 mm/min at room temperature. The data analysis was evaluated in tensile strength, %elongation, and modulus at break.

### 2.3.4 Biodegradability

Biodegradation test of the bio-composite film was performed by burying the sample into the soil for 3, 6, 9, and 12 months. The film specimen has 3 x 3 cm with 3 pieces was used in this test. The sample was buried in the soil with a high 10 cm from the ground and heap the soil cover the sample with a high of 5 cm. The soil has a humidity range of 70-80 % RH. The biodegradability was evaluated by the weight loss of the sample every 3 months for 12 months. The weighing was determined with a precision of 0.0001 g. The percentage of weight loss was calculated as follows.

$$\% \text{Weight loss} = \frac{W - W_0}{W} \times 100$$

Where:  $W$  = Weight of sample after burying in the soil (g)

$W_0$  = Weight of sample before burying in the soil (g)

### **3. The bio-polymer blend film from poly(lactic acid) incorporated with polybutylene succinate**

#### **3.1 Preparation of the bio-polymer blend film**

The bio-polymer blend film was prepared into 2 steps. The first step was to prepare the bio-polymer blend compound. The second step is the production of the bio-polymer blend film.

##### **3.1.1 The bio-polymer blend compound**

The bio-polymer blend compound was performed from poly(lactic acid); PLA incorporated with the content of polybutylene succinate; PBS by a twin-screw extruder. The raw materials were removed moisture by the universal oven at 60 °C for 2 hr before compound process. The condition for compounding at temperature 100, 120, 140, 150, 160, 170, 180, 180, 180, and 170 °C with screw speed 100 rpm. The ratio of the bio-polymer blend compound between PLA and various PBS was expressed in Table 5.

##### **3.1.2 The bio-polymer blend film**

The bio-polymer blend compound pellets from a twin-screw extruder were to production in the bio-polymer blend film. The blow film extruder was used for film production. The process condition has used the temperature as 150, 170, 180, and 170 °C with screw speed 90 rpm. The thickness and width of bio-composite film are 60-70 microns and 17 cm, respectively.

**Table 5 The ratio of the bio-polymer blend film between poly(lactic acid) and various contents polybutylene succinate**

Bio-polymer blend	Weight (g)	
	PLA	PBS
Neat PLA	100	0
L90_B10	90	10
L80_B20	80	20

### 3.2 Characterization of the bio-polymer blend compound

The bio-polymer blend compound was determined into 2 properties. The characterization was analyzed into morphology and the melt flow index. The detail can see in as following.

#### 3.2.1 Morphology

The cross-section morphology of the pellet of the bio-polymer blend compound from a twin-screw extruder was studied by a scanning electron microscope. The condition and detail can see as follows in 2.2.1.

#### 3.2.2 Melt flow index (MFI)

Melt flow index of the bio-polymer blend compound was tested using ASTM D1238E. The instrument and condition were presented in 2.2.2.

### 3.3 Characterization of the bio-polymer blend film

The bio-polymer film was determined into 4 properties. The characterization was studied the physical properties, chemical properties, mechanical properties, and thermal properties. The detail can be seen as follows.

#### 3.3.1 Physical properties

The cross-section of the bio-polymer blend film from tensile testing was used to evaluate the morphology by scanning electron microscope. The preparing of the sample can be seen in 2.3.2. The magnification has investigated at 1000X.

### 3.3.2 Chemical properties

The bio-polymer blend film has identified the crystalline and amorphous by X-ray diffraction (XRD). Using X-ray diffractometer (BRUKER, D2 Phaser; The Bruker®) for identity. The XRD spectra were determined by scanning from 5° to 40° and the scanning rate is 2°/min.

### 3.3.3 Mechanical properties

Tensile testing was used to explain the mechanical properties of the bio-polymer blend film. Tensile testing was characterized following ASTM D638 by using the universal testing machine for testing. The condition has used the same with the bio-composite film, which the detail was observed in 2.3.3. The analysis was considered in tensile strength, %elongation, and modulus at break.

### 3.3.4 Thermal properties

Thermal properties of bio-polymer blend film was tested by differential scanning calorimeter; DSC (METTLER TOLEDO, United States) under nitrogen (N<sub>2</sub>) atmosphere. Preparing the sample with weight around 8–10 mg. The conditions of DSC that first heated from room temperature to 200 °C with a heating rate of 10 °C/ min and then cooled down to 25 °C at a heating rate 10 °C/ min. The second heating was heated from 25 °C to 200 °C. The DSC thermograms were determined the glass transition temperature (T<sub>g</sub>), melting temperature (T<sub>m</sub>), and cold crystallization temperature (T<sub>cc</sub>). Moreover, calculation the degree of crystallinity (%X<sub>c</sub>) as follows.

$$\%Crystallinity = \frac{\Delta H_m}{W \cdot \Delta H_{m,PLA}} \times 100$$

Where:  $\Delta H_m$  = Enthalpy of the melting peak in endothermal process (J/g)

$\Delta H_{m,PLA}$  = Enthalpy of the melting peak of pure PLA (93 J/g)

$\Delta H_{m,PBS}$  = Enthalpy of the melting peak of pure PBS (210 J/g)

W = Weight fraction of each polymer in the bio-polymer (g)

#### **4. The bio-composite film from the bio-polymer blend incorporated with lemongrass leaf powder**

##### 4.1 Preparation of the bio-composite film

The bio-composite film has performed the bio-polymer blend in the ratio between PLA: PBS as 90: 10 incorporated with lemongrass leaf powder. The production of the bio-composite film has 2 steps. The first step is compounding of the bio-polymer blend and lemongrass leaf powder. The second step is the production of the bio-composite film.

##### 4.1.1 The bio-composite compound

The bio-composite compound was prepared from poly(lactic acid) and polybutylene succinate at 10 %w/w. Moreover, adding lemongrass leaf powder as natural filler was varied from 0, 2.5, 5.0, 7.5, and 10.0%w/w. The raw materials were removed moisture by the universal oven at 60 °C for 2 hr before compound process. Using a twin-screw extruder for compounding with temperature as 100, 120, 140,150, 160, 170, 180, 180, 180, and 170 °C with 100 rpm as screw speed. The ratio for compounding was exhibited in Table 6.

##### 4.1.2 The bio-composite film

The bio-composite compound from a twin-screw extruder was extruded and cut into a small pellet. The pellet was exported to produce the film by using the blow film extruder. The blow film was processed at temperatures 150, 170, 180, and 170 °C with a screw speed of 90 rpm. The thickness and width of bio-composite film are 60-70 microns and 17 cm, respectively.

**Table 6 The ratio of the bio-composite film between poly(lactic acid) and polybutylene succinate with various the content of lemongrass leaf powder**

Bio-composite	Weight (g)		
	PLA	PBS	LE powder
B10_LE2.5	87.5	10	2.5
B10_LE5.0	85.0	10	5.0
B10_LE7.5	82.5	10	7.5
B10_LE10.0	80.0	10	10.0

#### 4.2 Characterization of the bio-composite compound

The bio-composite compound was to test into 2 properties. The bio-composite film was characterized into morphology and the melt flow index. The detail can be seen as following.

##### 4.2.1 Morphology

The cross-section morphology of the pellet of the bio-composite compound from a twin-screw extruder was studied by a scanning electron microscope. The condition and detail can be seen as follows in 2.2.1.

##### 4.2.2 Melt flow index (MFI)

Melt flow index of the bio-composite compound was tested using ASTM D1238E. The instrument and condition were presented in 2.2.2.

#### 4.3 Characterization of the bio-composite film

The bio-composite film properties derived into 6 parts. The characterization studied the physical properties, chemical properties, mechanical properties, thermal properties, biodegradability, and field test. The detail can be seen as following.

##### 4.3.1 Chemical properties

Fourier Transform Infrared Spectroscopy (FT-IR): Perkin Elmer model Spectrum GX using for determining the functional group of the composite film. The bio-polymer blend film was assay with ATR mode and scanned in the wavenumber from 4000 – 400  $\text{cm}^{-1}$ .

X-ray diffraction (XRD): The bio-composite film has identified the crystalline and amorphous by X-ray diffraction (XRD). The diffractogram were determined by scanning from  $5^{\circ}$  to  $40^{\circ}$  and the scanning rate is  $2^{\circ}/\text{min}$ .

#### 4.3.2 Physical properties

Morphology: The surface and the cross-section of the bio-polymer film from tensile testing was used to evaluate the morphology by scanning electron microscope. The preparing of the sample can be seen in 2.3.2. The surface morphology was determined with magnification at 200X. The cross-section morphology of the bio-composite film was investigated at 1000X.

Water vapor permeability (WVP): The bio-composite film was assayed the water vapor transmission (WVTR) following ASTM E96-80. Which the detail was explained in 2.3.2.

Color parameter: The bio-composite film was measured the color parameter by using a calorimeter (Konica Minolta model CR-20) following ASTM E313-96. The processing was assigned in 2.3.2. The data report was exported the parameter consists of  $L^*$ ,  $a^*$ ,  $b^*$ , and  $\Delta E$ .

UV transmittance: The bio-composite film was measured %UV transmittance via UV-Vis spectrophotometer (specord 210 plus). Which the condition can observe in 2.3.2.

#### 4.3.3 Mechanical properties

Tensile testing was explained the mechanical properties of the bio-composite film. Tensile testing was characterized following ASTM D638 by using the universal testing machine for testing. The conditions are the same as described in 2.3.3. The analysis was considered in tensile strength, %elongation, and modulus at break.

#### 4.3.4 Thermal properties

Thermal properties of bio-polymer film was tested by differential scanning calorimeter under nitrogen ( $N_2$ ) atmosphere. The detail of preparing sample and condition was seen in 3.3.4. DSC thermograms were determined the glass transition temperature ( $T_g$ ), melting temperature ( $T_m$ ), and cold crystallization temperature ( $T_{cc}$ ). Moreover, study the degree of crystallinity of the bio-composite film.

#### 4.3.5 Biodegradability

The bio-composite film was embedded in soil for 1 and 2 months. The sample preparation and the calculation of percentage of weight loss can be seen in 2.3.4.

#### 4.3.6 Field test

The bio-composite film was covered the soil during environment for 1 and 2 months. This test took place at Naresuan University, Phitsanulok, Thailand, using real world conditions used for agriculture crops. The sample film covered the soil from November 2020 to January 2021 which is classified as the winter period of Thailand. The weather during this period recorded a low temperature of 18°C and the highest temperature was 28°C. The relative humidity was approximately 70-80%RH. The crops produced had a size of 25 x 50 cm per sample film. The specimen of bio-composite film was characterized by the physical properties (color parameter and UV transmittance) and mechanical properties (Tensile testing). The characterizations are described according to 2.3.2 in which color parameter measurement, UV transmittance, and tensile testing in 2.3.3, respectively.



## CHAPTER IV

### RESULTS AND DISCUSSION

This research has been interested in preparation and characterization of the mulch film from the biopolymer-based incorporated with the natural fillers. Poly(lactic acid); PLA and Polybutylene succinic acid; PBS were chosen as biopolymer-based. Ginger and lemon grass were chosen to use as natural filler. For ginger can be separated into 2 stages including early immature ginger and mature ginger. The mulch film manufactory was prepared in 2 steps, the first step a twin-screw extruder was used to prepare a compound, second step blow film extruder was used to blow a film. The investigation of PBS effect and natural fillers content on physical properties, chemical properties, mechanical properties, thermal properties, and biodegradability was studied.

The results are separated into 4 sections. The first section, characterization of natural fillers. The second, the effect of natural fillers into the PLA bio-composite film was studied then the good natural filler was chosen for improving the bio-composite film. The third section, the variation of PBS content in the PLA matrix was investigated to observe including physical and mechanical properties. The last section, the effect of different lemongrass leaf and PBS content was studied in the bio-composite film on the physical properties, chemical properties, mechanical properties, thermal properties, and biodegradability.

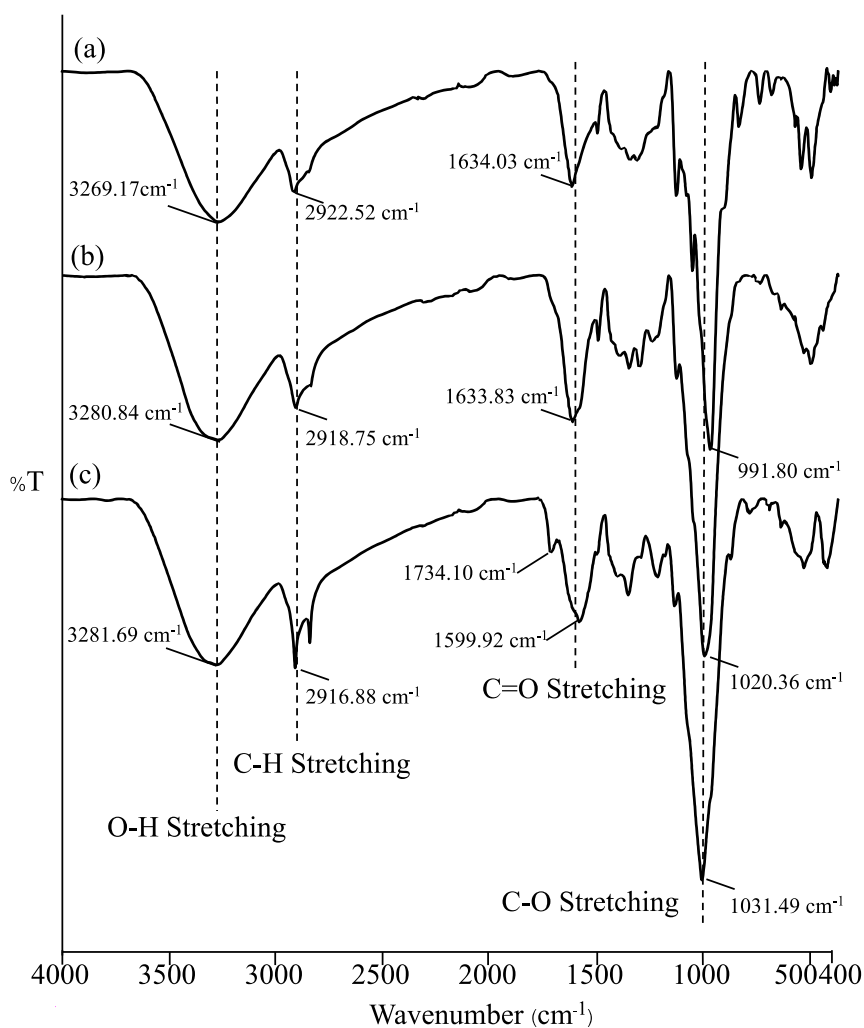
#### **The characterization of natural fillers**

In this research, we used 2 natural fillers, one was ginger which was used 2 stages consist of early immature ginger (EG) and mature ginger (MG), the other is lemongrass leaf (LE). All of natural fillers were prepared in powder with particle size around 45 microns. The one reason of choosing these 2 natural fillers due to they are the herb in Thailand, which are easier to find. Moreover, these natural fillers are non-toxic, non-contaminated to environment and biodegradable. The characterization of natural fillers, chemical properties by Fourier Transform Infrared Spectroscopy

(FT-IR), physical properties of the surface morphology natural fillers by Scanning electron microscopy (SEM) and the fiber length analysis of natural filler by using ImageJ software. The detail shows as following.

### **1. The chemical properties of the natural fillers**

All of the natural fillers were analyzed the functional group using Fourier Transform Infrared Spectroscopy (FT-IR) with ATR-mode in the wavenumber ranges on 400-4000  $\text{cm}^{-1}$ . These samples of natural filler were prepared to powder with particle size of approximately 45 microns. From the FT-IR spectra (Figure 26), four main peaks including O-H stretching (3200-3500  $\text{cm}^{-1}$ ), C-H stretching (2800-2900  $\text{cm}^{-1}$ ), C=O stretching (1500-1700  $\text{cm}^{-1}$ ), and C-O stretching (1000-1100  $\text{cm}^{-1}$ ) were observed. These bands found in natural fillers related to the functional group of cellulose, hemicellulose, and lignin. (56), which are previously reported as the chemical compositions of natural fillers (EG, MG, and LE). (54, 57) Among three natural fillers, LE powder have a slight different band of FT-IR at 1734.10  $\text{cm}^{-1}$  as seen in Figure 26c and this result corresponded to the study of Lemma Dadi Bekele and co-worker. The band at about 1734.10  $\text{cm}^{-1}$  was proposed as the result of C=O stretching of the acetyl group and linked to the ester linkage of carboxylic group of ferulic acid and *p*-coumaric acid of lignin. (54)



**Figure 26 FT-IR spectra of (a) early immature ginger powder, (b) mature ginger powder, and (c) lemongrass leaf powder**

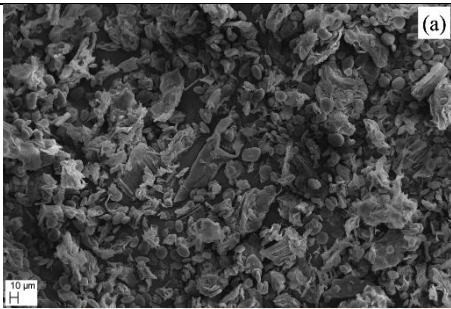
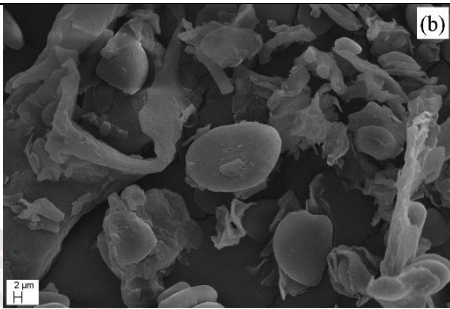
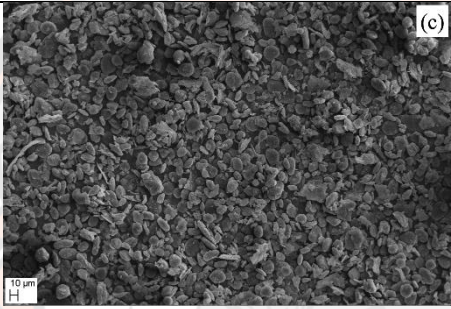
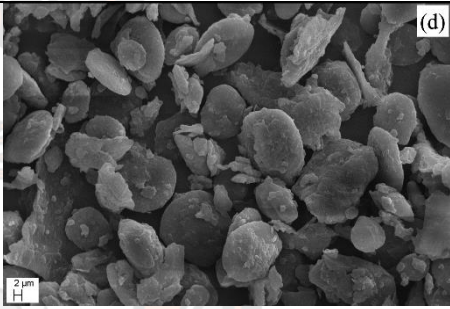
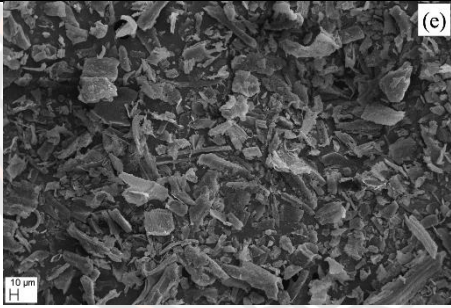
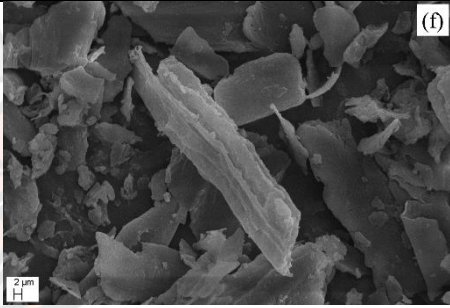
## 2. The morphology of the surface natural fillers

The morphology was observed by scanning electron microscopy (SEM). These samples of natural filler were prepared to powder with particle size approximately 45 microns. To study the morphology of each surface natural fillers.

From the SEM images show the characteristic of each of the natural fillers. Table 7a presents the characteristic of early immature ginger particles at magnification 200X. The particles are flakes and flat circle with distribution of the different fiber length. Table 7b presents the morphology of early immature ginger at

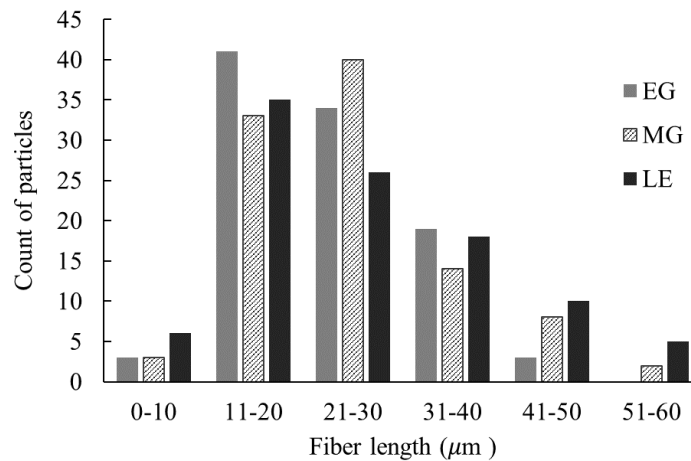
1000X that the surface morphology has a smooth surface. Table 7c shows the morphology of mature ginger at 200X, which the particles are flakes like the early immature ginger particles. Table 7d presents the morphology of mature ginger at the magnification 1000X that the surface morphology shows the obviously roughness more than the early immature ginger powder surface. The surface of ginger particles is covered with another layer of hemicellulose and lignin and other substances. The roughness surface of the mature ginger particles may causing from the maturation of ginger due to the increasing of maturation stage influences the fiber in the ginger. The other research studies the stage of ginger affects the crude fiber in ginger. It was found the early immature ginger has a crude fiber of 1.35 g/100g. While the mature ginger has increased the crude fiber to 1.52 g/100g. (47) The increasing of fiber influences increasing the amount of hemicellulose and lignin in mature ginger too. The percentage of cellulose, hemicellulose, and lignin were reported as 70.05%, 26.45%, and 3.5%, respectively. (57) Table 7e exhibits the lemongrass leaf particles at magnification 200X that the most particles show flakes and different shapes. Some particles reveal a rod shape and irregular shape, which related to the fiber characteristic of lemongrass leaf. The lemongrass leaf is a monocotyledon, which it has a long fiber. (58) Table 7f shows the morphology of the lemongrass leaf particles that these particles have a rod shape and flake with a rough surface. Lemongrass leaf has the chemical compositions as cellulose, hemicellulose, and lignin in 39.5%, 22.6%, and 28.5%, respectively. (54)

**Table 7** The morphology of natural fillers

Sample	Magnification	
	200X	1000X
EG powder		
MG powder		
LE powder		

### 3. Fiber length of natural fillers

The natural fillers were prepared under sieve size 45 microns thus the particle of natural fillers was varied in different length. The fiber length was studied by ImageJ software. The result was reported in average length of each natural filler.



**Figure 27 The fiber length of each natural fillers**

Figure 27 proposes the distribution of fiber length for each of natural fillers. Each of the natural fillers has a fiber length from 10 to 40 microns that these particles can observe in SEM images in Table 7. For some particles have a length more than 45 microns due to the during process of measurement in length of natural fillers, which LE powder has the rod shape and flake thus affects to the fiber length more than 45 microns. The most length of early immature ginger and mature ginger has been distributed in 11-30 microns. While the fiber length of lemongrass leaf powder has broad range of fiber length from 11-40 microns. The different length and shape of natural fillers affect to the mechanical properties of the bio-composite. In the theory of fillers, the shape, size, and surface morphology of fillers influences the particle distribution in the polymer matrix. (41)

In the summary, the morphology of each natural filler has a different shape, roughness of a surface, and fiber length. Both stages of ginger particles have a flat circle shape that the surface morphology of mature ginger has roughness more than early immature ginger. The length of fiber has a narrow range from 11-30 microns.

While the lemongrass leaf particles have an irregular shape and vary broadly the fiber length from 11-40 microns.

### **The effect of each natural filler in the bio-composite base on PLA matrix**

The purpose of this section is studied the effect of natural filler to bio-composite on the chemical properties, physical properties, mechanical properties, and biodegradability. The natural fillers were varied at 0, 2.5, 5.0, and 7.5 %w/w. These results consist of 2 sections bio-composite compound and bio-composite film. The acronym of bio-composite in this part consists of a letter such as “LE” that means the type of natural filler following a number of the percentage of natural filler. The acronym illustrates as following in Table 8.

**Table 8 The acronym of bio-composite compound and film**

<b>Natural filler</b>	<b>Content of natural filler (%w/w)</b>	<b>Acronym</b>
Non-filler	0	PLA
Early immature ginger	2.5	EG_2.5
	5.0	EG_5.0
	7.5	EG_7.5
Mature ginger	2.5	MG_2.5
	5.0	MG_5.0
	7.5	MG_7.5
Lemongrass leaf	2.5	LE_2.5
	5.0	LE_5.0
	7.5	LE_7.5

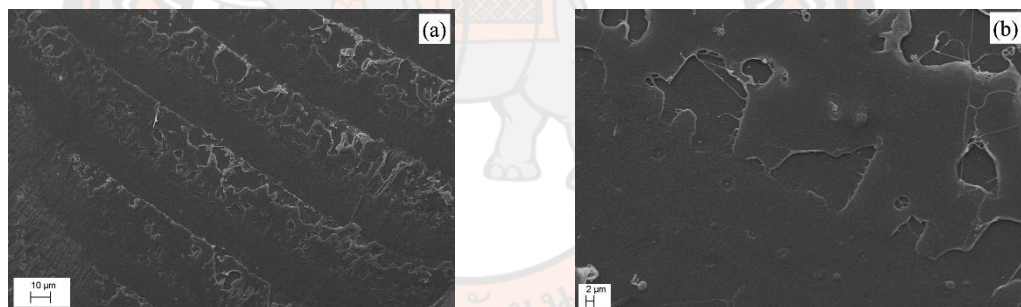
#### **1. Bio-composite compound from PLA and natural fillers**

The sample of the bio-composite compound was performed by a twin-screw extruder. To study the distribution of natural fillers in the polymer matrix via morphology using scanning electron microscopy (SEM). Furthermore, the melt flow index (MFI) of the bio-composite compound was studied.

### Morphology of bio-composite compound

To study the distribution with the same content of natural fillers (5% natural filler) in the PLA matrix, which was studied by the cross-section of bio-composite compound from pellet cutting by the twin-screw extruder. The analysis of the different types of natural fillers affects to the adhesion between fillers and matrix. This bio-composite compound was investigated by scanning electron microscopy (SEM) at magnification 200X for observing the overall cross-section morphology and natural filler distribution. At magnification 1000X for expressing of the interaction between natural filler and polymer matrix.

Figure 28-31 are indicated the cross-section morphology of the PLA compound and bio-composite compound incorporated with the different types of natural fillers. The cross-section of the neat PLA compound is shown in Figure 28a-b that the morphology has a clear surface.

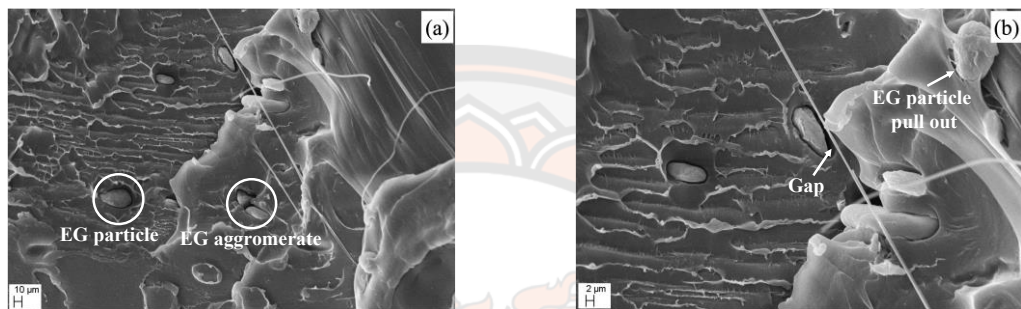


**Figure 28 Cross-section morphology of PLA compound at magnification (a) 200X, and (b) 1000X**

The addition of 5% natural fillers into the polymer matrix shows the natural fillers distribute and bind in the polymer matrix, but some particles pulled out of the polymer matrix. The bio-composite compound incorporates with the early immature ginger powder that exhibited in Figure 29a-b. The EG particles distribute into the polymer matrix and some particles pull out of the polymer matrix which generates the agglomerate in the polymer matrix in Figure 29a. The agglomerate of EG particle in polymer matrix cause from the surface morphology and length fiber. The EG particles have a smooth surface and small particles that easy to aggregates and tack of the

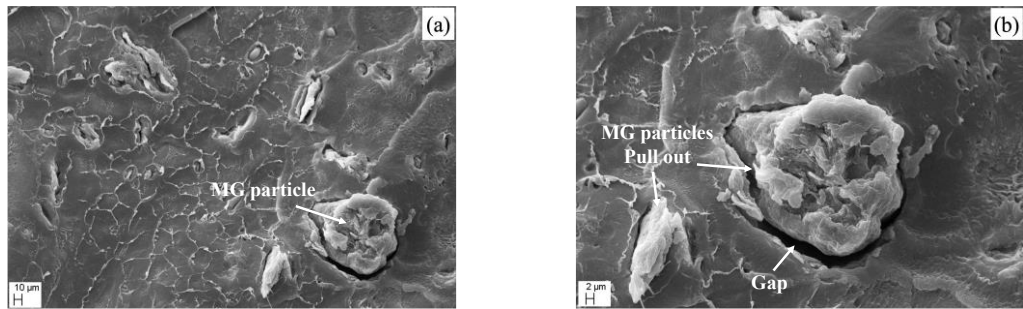


particles. The EG powder has the chemical composition as cellulose which explains the tack of particle. The cellulose has a hydrophilicity due to increasing molecular interaction of fiber that causing to generate the EG agglomerate in the polymer matrix. Figure 29b shows the interaction between EG particles and polymer matrix, which expresses the gap between particles and matrix. The agglomerate of the EG particles affects the film surface that has rough surface.



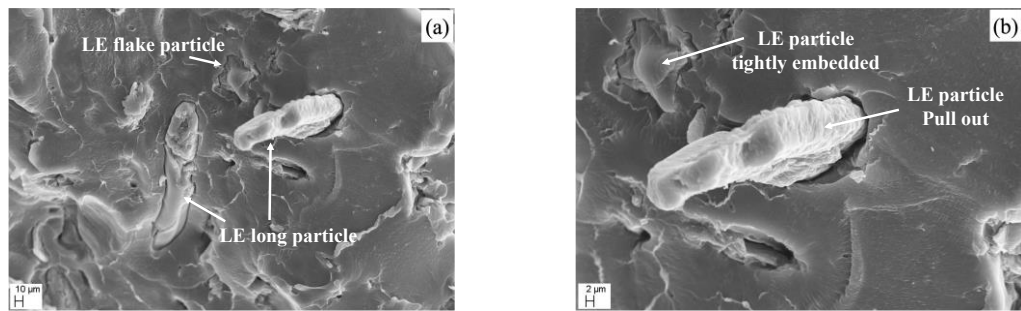
**Figure 29 Cross-section morphology of bio-composite compound incorporated with early immature ginger powder at magnification (a) 200X, and (b) 1000X**

The cross-section morphology of the bio-composite compound with mature ginger powder presents in Figure 30a-b. It was found the MG particles distributed in the polymer matrix. The MG particles distributed and embedded in the polymer matrix. The rough surface of MG particles helps to good adhesion between particles and matrix. However, the large particle size reduced the adhesion with the matrix. The large particle pulled out of the polymer matrix and the morphology has gap between MG particles and matrix.



**Figure 30 Cross-section morphology of bio-composite compound incorporated with mature ginger powder at magnification (a) 200X, and (b) 1000X**

The cross-section morphology of the bio-composite with lemongrass leaf powder expresses in Figure 31a-b. The LE particles were distributed as random orientation and tightly imbed in the polymer matrix. The adhesion area between LE particles and polymer matrix indicates in Figure 32b, it was found the gap between LE particle and matrix that was less than the EG and MG particles gap. In the meanwhile, it was found the LE particles were pulled out of the matrix because of the LE particles have a long particle more than ginger that affects pulling out of the matrix during the cutting to the small pellet from rod compound by a twin-screw extruder. The morphology of bio-composite compound incorporated with LE particle was indicated the rough surface of LE particles which has a good adhesion more than ginger. LE particles have flake, rod particles and roughness. The polymer chain can be attached and combined with the rough surface of LE particle that affects to the interaction between particles and matrix are hence. This good adhesion of morphology influences the properties of the bio-composite.



**Figure 31 Cross-section morphology of bio-composite compound incorporated with lemongrass leaf powder at magnification (a) 200X, and (b) 1000X**

The natural fillers contain the cellulose, hemicellulose, lignin, and another substance on the surface. (47, 56, 57) The substances on the surface are the hydrophilic compound which has more intermolecular force effect that it is easy to tack and aggregates between particles.

In the summary, the addition of natural fillers into the PLA matrix, the particles of natural filler were distributed in the polymer matrix and reduced the continuous phase of the polymer matrix. The rough surface of the natural filler helps to the particles and matrix has a good adhesion.

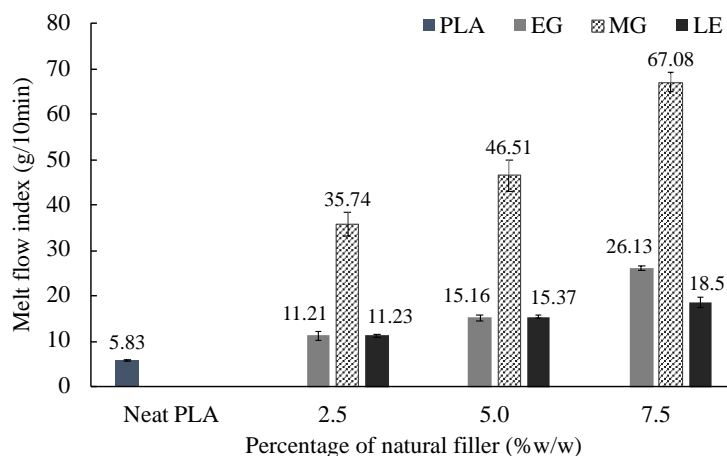
#### **The effect of natural fillers on melt flow index (MFI)**

The melt flow index of the bio-composite compound incorporated with different natural fillers is attributed in Figure 32. The result shows the neat PLA has MFI of 5.83 g/10 min. The addition of natural fillers into the PLA matrix exhibits increasing the MFI more than the neat PLA compound (>5.83 g/10 min). Moreover, the increasing percentage of natural filler affects to the MFI increasing. The reason for this behavior causes from the natural fillers distribution in the polymer matrix that it reduces the continuous phase of the polymer matrix. Moreover, the addition of natural filler affects the reducing entanglements of polymer chain cause to the increasing of MFI. The MFI influences the melting in the blow film manufactory process and controls the temperature for the processing. Furthermore, MFI can predict miscibility

of the bio-composite that the high MFI affects to the immiscibility of the bio-composite.

MFI of bio-composite compound ranges on 11.00-27.00 g/10 min was incorporated with EG powder. The addition of MG powder into the PLA matrix jumped up to 35.00-67.00 g/10 min. The most fiber of MG has a length approximately 21-30 microns, which is non-various length cause poor MG distribution and generate agglomeration. It affects to the interaction between polymer chain and filler that has poor adhesion, which shown gap between MG particle and polymer that seen in SEM image in Figure 30.

The incorporation of lemongrass leaf powder in the bio-composite films has the MFI around 11.00-19.00 g/10 min, which is a slightly different from the neat PLA. Due to the lemongrass powder is well distributed in the PLA matrix cause the MFI slightly changed. The SEM image in Table 7e-f shows the surface morphology of LE particles that has a roughness more than ginger. Thus, the interaction between LE particle and polymer chain is well adhesion more than the bio-composite compound with ginger. Furthermore, the LE particles have various fiber lengths, and different shapes therefore the LE particles can well distribution in a random orientation that according to the SEM image and the fiber length results. The LE particles show well distribution in the polymer matrix and have higher miscibility than the bio-composite compound incorporated with ginger.



**Figure 32 Melt flow index of bio-composite compound incorporated with each natural fillers and different content of natural fillers**

## 2. Bio-composite film from PLA and natural fillers

The bio-composite compound pellets from a twin-screw extruder were produced the bio-composite film by blow film extruder with 70-80 microns as thickness and width of 16-17 cm. This part studies the effect of different types of natural fillers and various percentages of natural fillers added into the neat PLA film. The natural fillers were varied at 0, 2.5, 5.0, and 7.5 %w/w. To characterize the chemical properties (FT-IR), mechanical properties (Tensile testing), physical properties (SEM, Color parameter, UV-transmittance, and Water vapor permeability), and biodegradability of the bio-composite film. Moreover, the aim of this part is selection of the appropriated natural filler for developing and producing the mulch film. The acronym of the bio-composite films was assigned following in Table 8.

### The chemical properties of the bio-composite film

The determination of the functional group of the bio-composite film with adding natural filler and without adding natural filler into polymer matrix were analyzed in this section. All bio-composite films were assayed the functional group in the region ranges  $400\text{-}4000\text{ cm}^{-1}$  by using Fourier Transform Infrared Spectroscopy (FT-IR) with ATR-mode.

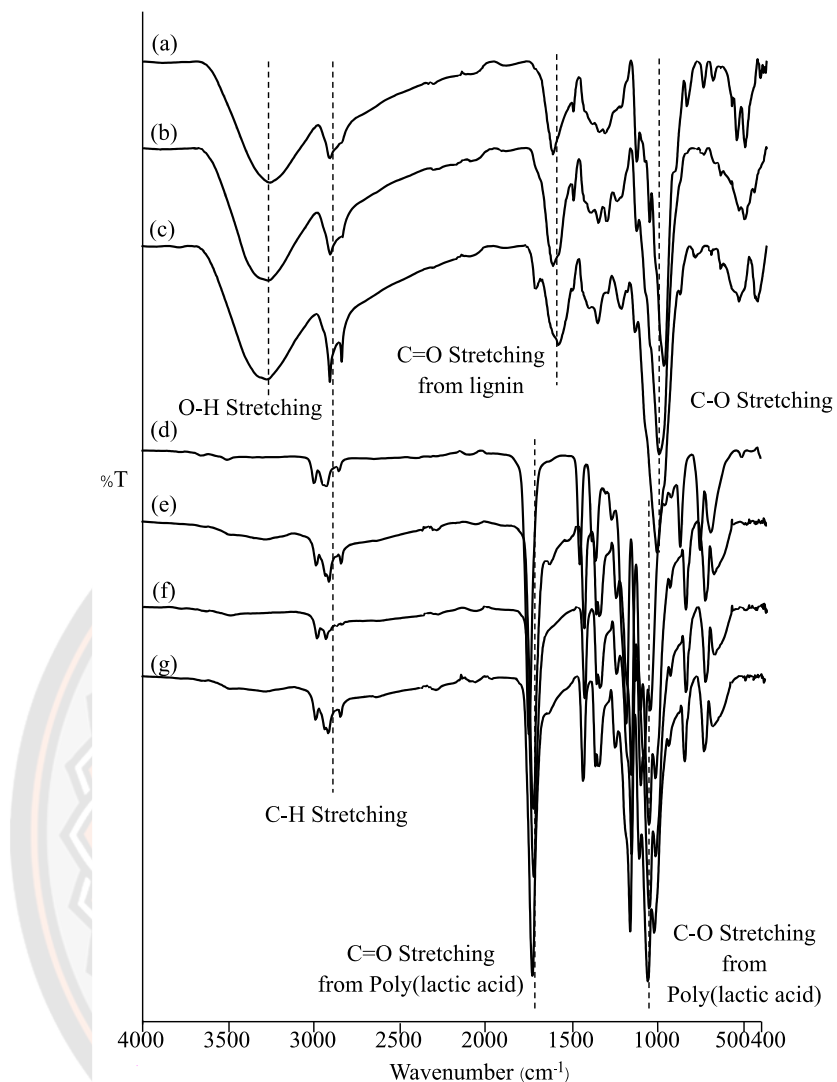
Figure 33 represents the FT-IR spectrum of natural fillers and the bio-composite film. The interpretation of the FT-IR spectrum of natural fillers and the bio-composite films incorporated with different types of natural fillers were presented in Table 9. All the natural fillers were indicated the main 4 peaks consist of O-H stretching at 3200-3300  $\text{cm}^{-1}$ , C-H stretching region on 2900-3000  $\text{cm}^{-1}$ , C=O stretching as 1600-1700  $\text{cm}^{-1}$ , and C-O stretching around 1000-1100  $\text{cm}^{-1}$ . These spectrums shown the functional groups according to the chemical composition of natural fillers as cellulose, hemicellulose, and lignin. The description of assign the functional group of natural fillers can clearly explain in the previous part of the chemical properties of natural fillers.

The neat PLA film exhibited the 3 main spectrums as C-H stretching at 2994.01  $\text{cm}^{-1}$ , C=O stretching has appeared 1745.66  $\text{cm}^{-1}$ , and at 1077.61  $\text{cm}^{-1}$  was interpreted C-O stretching. These functional groups from assign have related to the chemical structure of the poly(lactic acid). The addition of natural fillers included EG powder, MG powder, and LE powder it was found the FT-IR spectrum of the whole bio-composite films has a similar FT-IR spectrum with the neat PLA film. However, the FT-IR spectrum of the bio-composite film incorporated with natural fillers has a shift region as slightly. All of the bio-composite films were presented at the wavenumber ranges 2900-3000  $\text{cm}^{-1}$  were O-H stretching, C-H stretching has arrived on 1700-1800  $\text{cm}^{-1}$ , and C-O was emanated on 1000-1100  $\text{cm}^{-1}$ . The FT-IR spectrum of all bio-composite films only express the function group of the poly(lactic acid) structure and not different from the neat PLA spectrum. Moreover, the FT-IR spectrum did not appear the function group of the natural filler when the spectrum was observed that it did not find the peak of O-H stretching (Figure 33). Reasoning about not appear O-H stretching in the FT-IR spectrum of bio-composite films may be from the effect of the natural filler content in PLA matrix. The comparison of the ratio between the polymer matrix and natural filler was found that it has the amount of natural filler less than the whole PLA matrix. The result related to the SEM image in Figure 28-31, they represented the cross-section morphology of bio-composite compound that was found the area of PLA matrix has a larger than the area of natural fillers. Also, the analysis was characterized under ATR mode that is another reason because the ATR mode was characterized only the specimen surface. The addition of

natural fillers into the PLA matrix, the natural fillers were bound and embedded in the PLA matrix which non-pulled out from the surface of the bio-composite film. According to the SEM images in Table 11-13 which were exhibited the bio-composite film surface. Thus, the previous reasons affected the FT-IR spectrum and also not presented the O-H stretching in the bio-composite film.

**Table 9 FT-IR spectrum interpretation of natural fillers and the bio-composite film**

Samples	Wavenumber (cm <sup>-1</sup> )			
	O-H Stretching	C-H Stretching	C=O Stretching	C-O Stretching
Natural fillers				
EG powder	3269.17	2922.52	1634.03	991.80
MG powder	3280.84	2918.75	1633.83	1020.36
LE powder	3281.69	2961.88	1599.92	1031.49
Films				
Neat PLA film	-	2994.01	1745.66	1077.61
EG film	-	2921.48	1743.73	1077.86
MG film	-	2994.89	1745.03	1077.95
LE film	-	2922.40	1744.16	1077.88



**Figure 33 FT-IR spectrum of natural fillers as (a) early immature ginger powder, (b) mature ginger powder, (c) lemongrass leaf powder and the bio-composite film as (d) neat PLA film, (e) EG film, (f) MG film, and (g) LE film**



### **The morphology of the bio-composite film**

The bio-composite films were evaluated the morphology of the bio-composite films via Scanning electron microscopy (SEM). To evaluate the surface morphology and the cross-section morphology of bio-composite films after carries from tensile testing. To study the effect of the different types and different content of natural fillers on the morphology of the bio-composite film. The surface morphology of bio-composite films was determined at magnification 200X for analyzed the overall surface and at 1000X for investigated the characteristic of natural filler particles on the surface morphology. The cross-section morphology of bio-composite films was observed at magnification 200X for evaluated the morphology after the tensile testing. Moreover, it can observe the surface morphology in a high angle direction. For magnification 1000X can be used to analyze the interaction between natural fillers and polymer matrix of the specimen after carried from the tensile test.

Table 10a-b presents the surface morphology of the neat PLA film. It was found the surface morphology was clear and smooth. The surface area did not have voids. Table 10c shows the cross-section morphology of the neat PLA film, which observed in the high angle direction that exposed smooth surface and without appeared some particle pulled out of surface. At magnification 1000X can study the attribution of the cross-section morphology after received the tension force. The neat PLA film after carried the tension force shown the polymer was slightly extended, moreover the phase is not separated. The addition of natural fillers into the PLA matrix affected to change in the surface and cross-section morphology of bio-composite film. The incorporation of natural fillers may be effect on the attribution of bio-composite films.

**Table 10 Surface morphology and cross-section of the neat PLA film after carried the tensile testing**



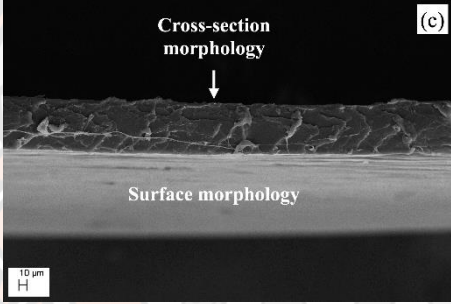
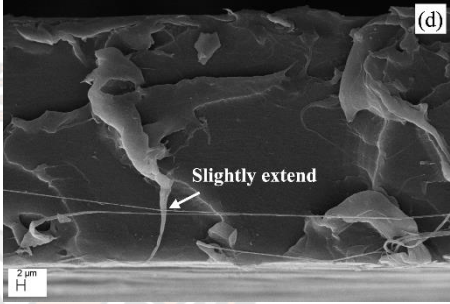
Sample	Magnification	
	200X	1000X
Surface of neat PLA		
Cross-section of neat PLA		

Figure 11 shows the surface morphology of the bio-composite films incorporated with the different content of early immature ginger powder (EG film). From the SEM image was found the surface morphology of EG particles was well distributed in the overall of the bio-composite film but the EG particle get the agglomerate in the bio-composite film. The addition of EG powder affected to roughness to the bio-composite films. Moreover, the increasing content of EG powder affected the EG particle has generated the agglomerate and causing of increasing roughness appearance. At the high angle direction in cross-section image showed roughness of the surface. The agglomerate and large EG particles were shown in Table 11b-d-f, that the EG particles were bulged and separated from the bio-composite film surface. Table 11 g-l presents the cross-section of the EG film after tensile testing. The cross-section of bio-composite film presented the distribution of EG particles in the polymer matrix and investigated the adhesion between EG particle and polymer matrix after carried on tension force. The cross-section of EG film after

the tensile test at magnification 1000X that are clearly presented the interaction during EG particle and polymer matrix. The specimen of EG film after received the tension force which affected to the PLA matrix has a slightly stretch and some particles of EG fallout from the matrix because the voids in the matrix. Some particles were bound and embedded in the polymer matrix that covered and involved by a polymer matrix. The interaction of EG particles and polymer matrix has a poor adhesion because the interaction areas have a gap, hole, and pulled out from the polymer matrix. The poor distribution of EG particles was reduced by the continuous phase of the polymer matrix that influences the mechanical properties of bio-composite film.

**Table 11 Surface morphology and cross-section of EG film after carried the tensile testing**

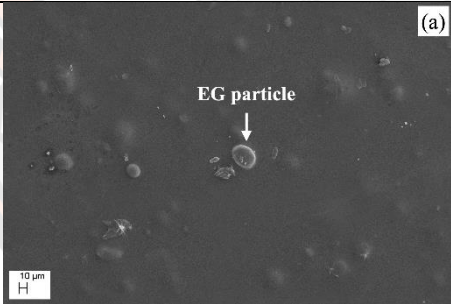
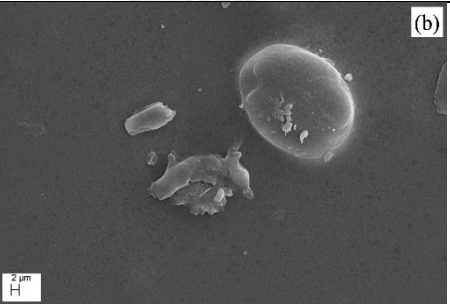
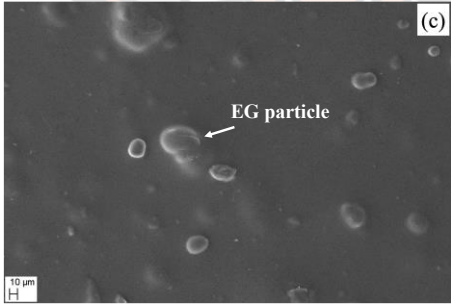
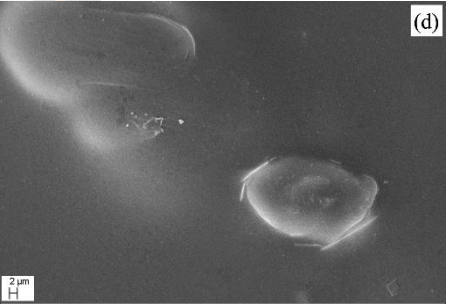
Sample	Magnification	
	200X	1000X
Surface of EG_2.5		
Surface of EG_5.0		

Table 11 (Cont.)

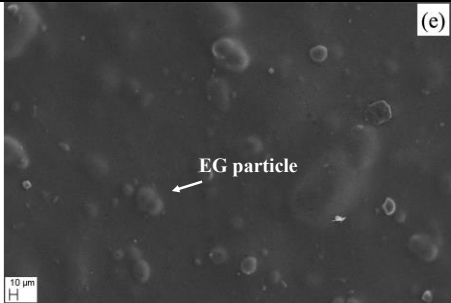
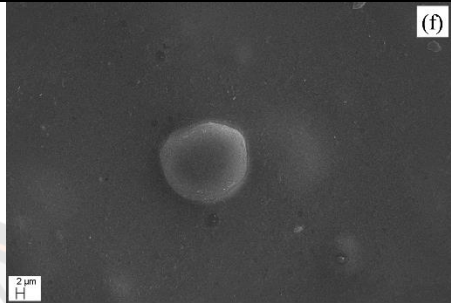
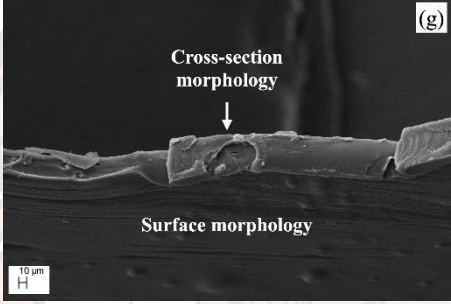
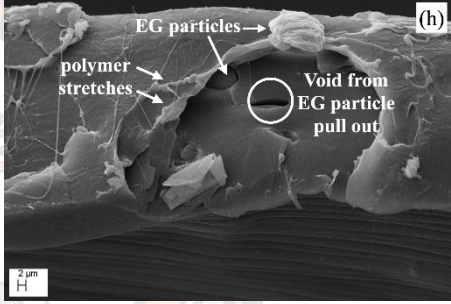
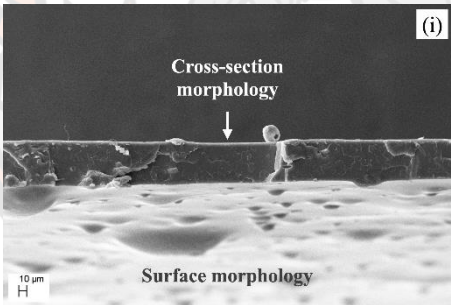
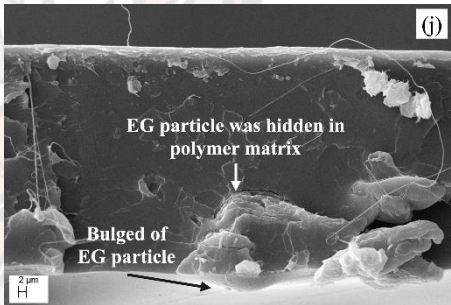
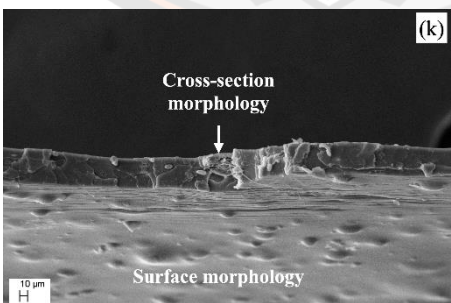
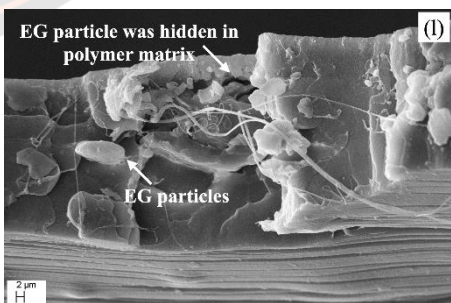
Sample	Magnification	
	200X	1000X
Surface of EG_7.5	 <p>(e)</p>	 <p>(f)</p>
Cross-section of EG_2.5	 <p>(g)</p>	 <p>(h)</p>
	 <p>(i)</p>	 <p>(j)</p>
Cross-section of EG_7.5	 <p>(k)</p>	 <p>(l)</p>

Table 12a-f exhibits the surface morphology of the bio-composite film when adding various content of mature ginger powder (MG film). It was found the distribution of MG particles in the bio-composite film when increasing the content of MG powder that increased more agglomerate in the polymer matrix. The MG powder was non-compatible with the PLA matrix because the addition of MG powder was separated from the PLA matrix causing the cracking between MG particles and the PLA surface. Looking in the high angle direction in Table 12g-l shows the cross-section of the bio-composite film after the tensile testing. It was found the surface of the bio-composite films has roughness and the agglomerated MG particles are bulge and pulled out of the bio-composite film. The interaction of MG particles and PLA matrix can study in the cross-section of the bio-composite films. The magnification at 1000X of SEM images shows the interaction of MG particle and PLA matrix when the specimen was received tension force, the PLA matrix was reduced stretching when compared with EG film. The MG particle embedded in the PLA matrix that the cross-section of the bio-composite has a less smooth surface causing an increase the more discontinuous phase.

From the result, the attribution of this MG film shown immiscible phase that may be affect to the mechanical properties of the bio-composite film. Both ginger powders were poorly distribution in the PLA matrix are might from the effect of the characteristic of particle. Both ginger powders have almost flat circle particles and lose the rough surface that may be causing the poor adhesion between particle and matrix. While the particle sizes have nearly size causing the ginger particle can be easier to generate the agglomerate in the PLA matrix and the distribution is less ability.

**Table 12 Surface morphology and cross-section of MG film after carried the tensile testing**

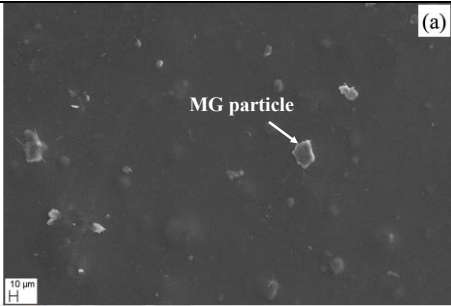
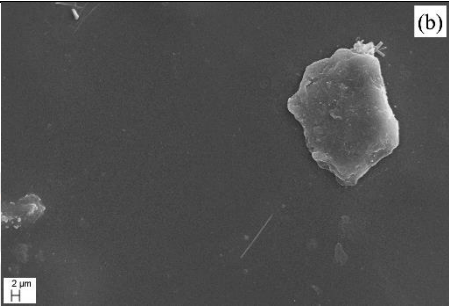
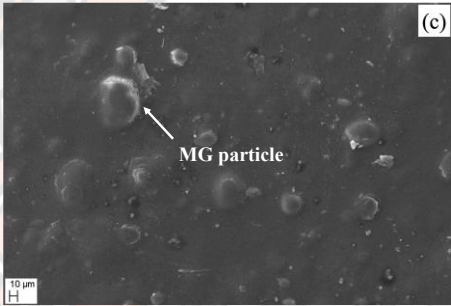
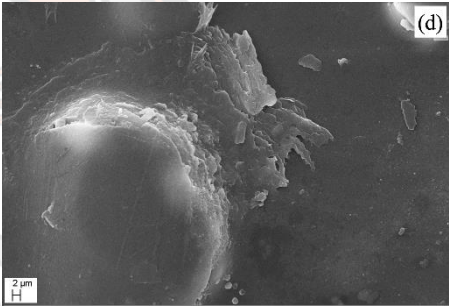
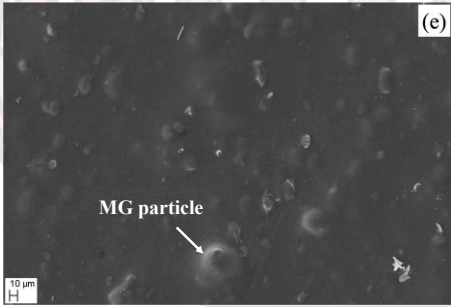
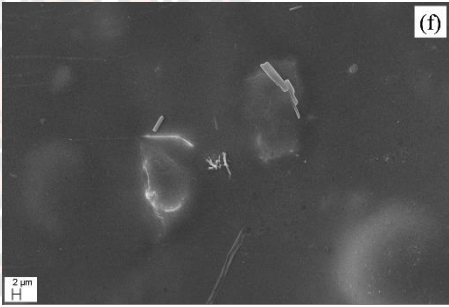
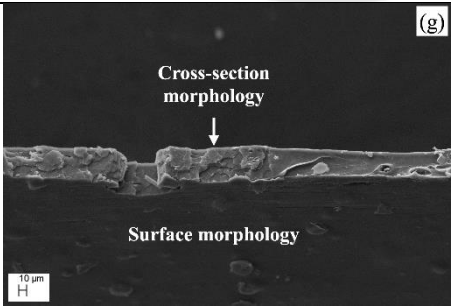
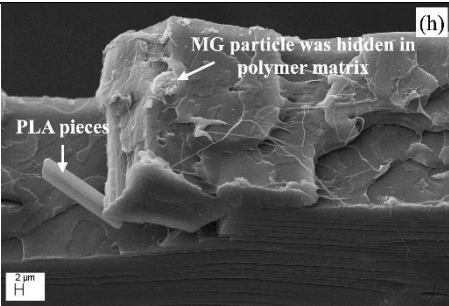
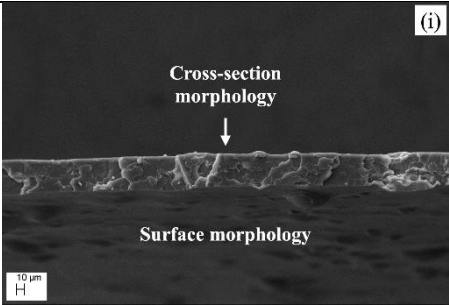
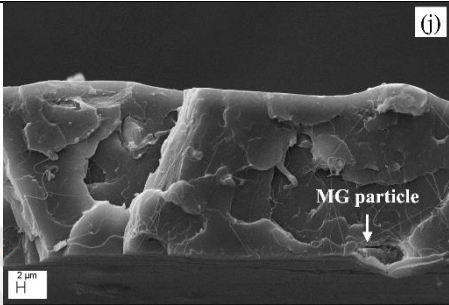
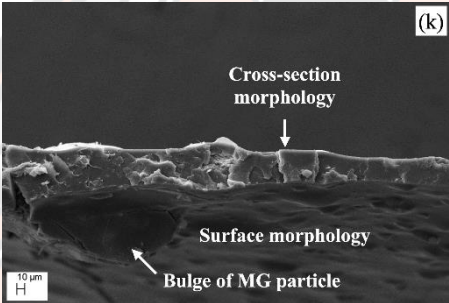
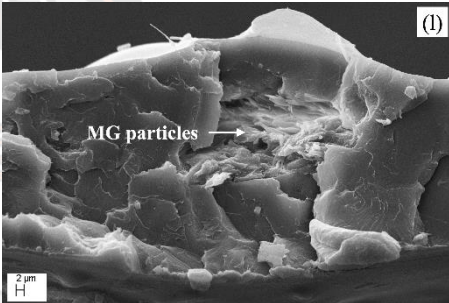
Sample	Magnification	
	200X	1000X
Surface of MG_2.5	 <p>(a)</p>	 <p>(b)</p>
Surface of MG_5.0	 <p>(c)</p>	 <p>(d)</p>
Surface of MG_7.5	 <p>(e)</p>	 <p>(f)</p>
Cross-section of MG_2.5	 <p>(g)</p>	 <p>(h)</p>

Table 12 (Cont.)

Sample	Magnification	
	200X	1000X
Surface of MG_5.0	 <p>(i)</p> <p>Cross-section morphology</p> <p>Surface morphology</p> <p>10 µm</p>	 <p>(j)</p> <p>MG particle</p> <p>2 µm</p>
Surface of MG_7.5	 <p>(k)</p> <p>Cross-section morphology</p> <p>Surface morphology</p> <p>Bulge of MG particle</p> <p>10 µm</p>	 <p>(l)</p> <p>MG particles</p> <p>2 µm</p>

The bio-composite film incorporated with LE powder (LE film) has shown the surface morphology in Table 13a-f and expressed the cross-section morphology after the tensile test as Table 13g-l. The surface morphology of the LE film has a rough surface when compared with the neat PLA film and increasing the content of LE powder causing roughness of the film surface. The LE particles were well distributed in the LE film that has seen in the SEM image the overall LE film surface has LE particle distribution. Since they have a different sizes and various shapes distributed on the surface that can be observed the characteristics as the bulge LE particle on the LE film surface. The addition of LE particles has a less convex from the PLA matrix when compared with both bio-composite films with ginger powder. The roughness of LE film has less than EG and MG films at the same content of natural filler adding. The different sizes and shapes of LE powder are a good attribute cause the LE particle can be well distributed and involved with the PLA matrix. However, the large particle size of LE powder affected to the roughness too, which can be observed in Table 13g that it shows the LE particle is convex from the PLA

matrix that may affect to some properties of the bio-composite film. The study of the interaction between LE particle and PLA matrix was analyzed in the cross-section of the bio-composite film after the tensile test can be seen in Table 13. The specimen was carried the tension force causing the LE film was cracked which the LE particle was pulled out from the PLA matrix but the area between the LE particle and the PLA matrix was non-separate, non-voids and holes. The LE particle was embedded in the PLA matrix and well adhesion with the PLA matrix due to the surface of LE particle has more roughness causing the PLA matrix can be easier to involve.

From the morphology results of the whole bio-composite film can be conclude the addition of natural fillers affects the morphology as reducing the continuous phase of the bio-composite film. The increasing content of natural fillers affects to the distribution in the PLA matrix and the surface of bio-composite film show more roughness when increasing the content of natural fillers. LE powder has well-distributed in the PLA matrix more than both ginger powders. Furthermore, the rough surface of the LE particle has contributed to the adhesion with the PLA matrix. However, the addition of all-natural fillers causing the immiscible bio-composite film.



**Table 13 Surface morphology and cross-section of LE film after carried the tensile testing**

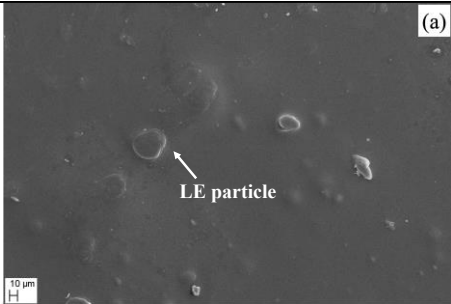
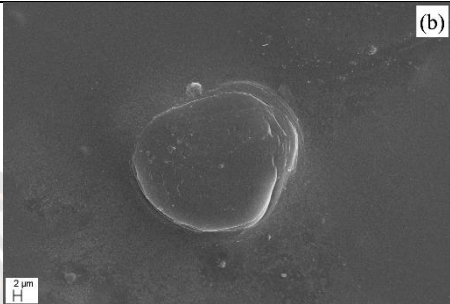
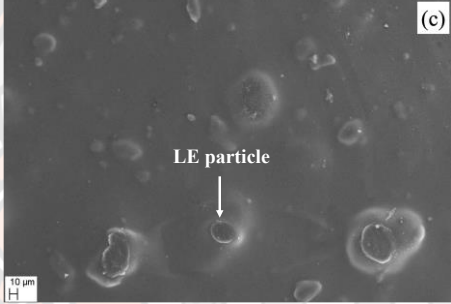
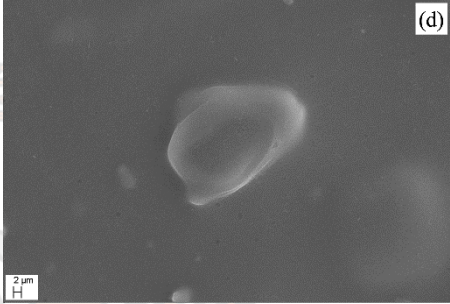
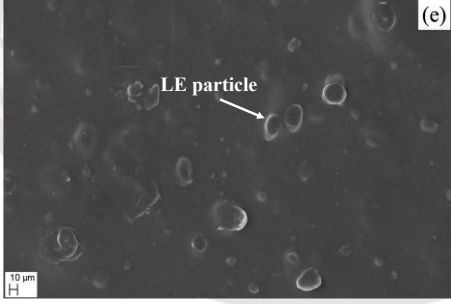
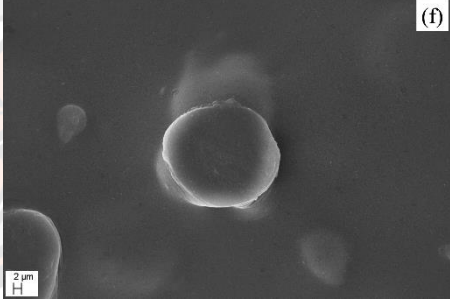
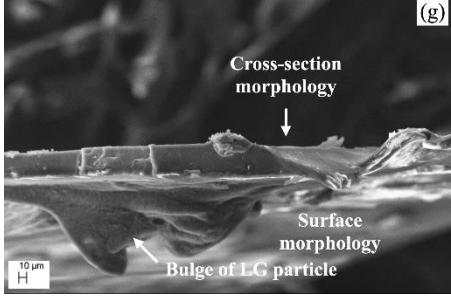
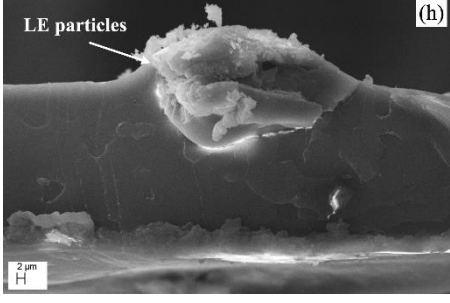
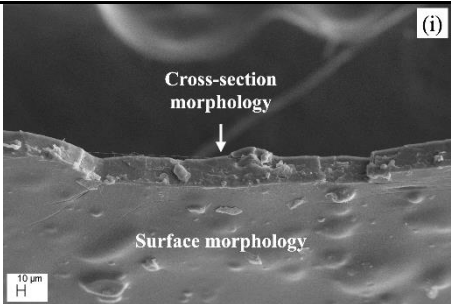
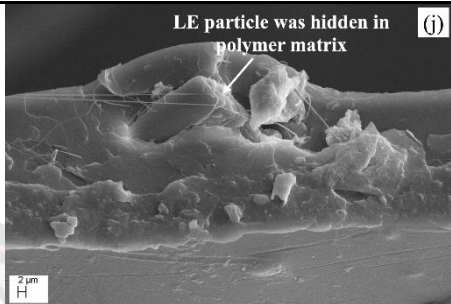
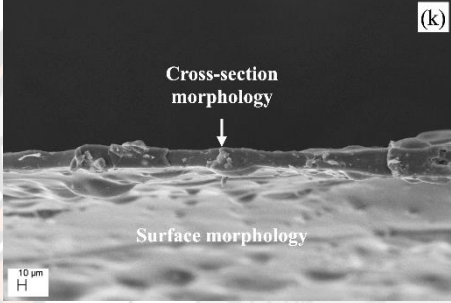
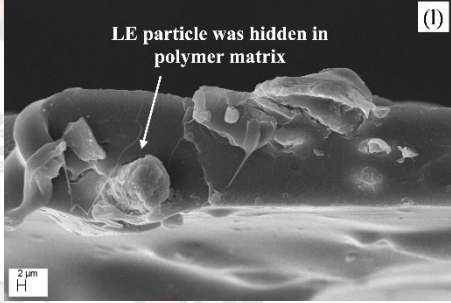
Sample	Magnification	
	200X	1000X
Surface of LE_2.5	 <p>(a)</p>	 <p>(b)</p>
Surface of LE_5.0	 <p>(c)</p>	 <p>(d)</p>
Surface of LE_7.5	 <p>(e)</p>	 <p>(f)</p>
Cross-section of LE_2.5	 <p>(g)</p>	 <p>(h)</p>

Table 13 (Cont.)

Surface	Magnification	
	200X	1000X
Cross-section of LE_5.0		
Cross-section of LE_7.5		

### The mechanical properties of the bio-composite film

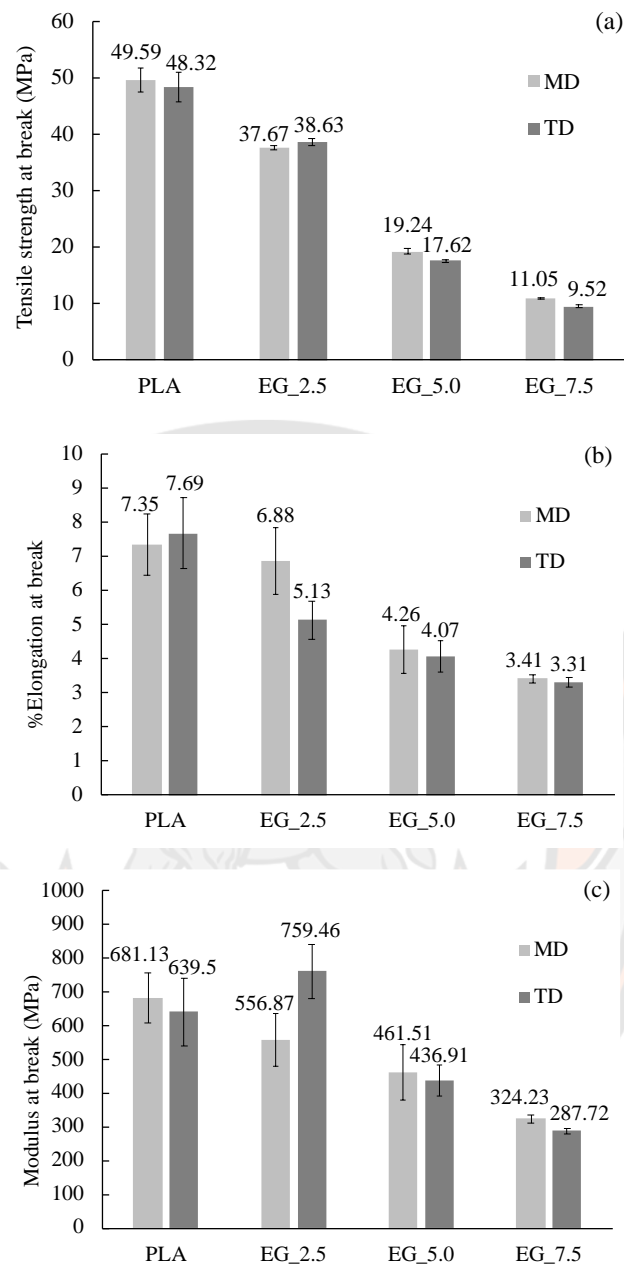
The bio-composite films were characterized the mechanical properties by tensile testing. The analysis was investigated the Tensile strength at break, %Elongation at break and Modulus at break. The neat PLA film and the bio-composite film were characterized the tensile test in mechanical direction (MD) and transverse direction (TD) due to the film process using by blow film extruder thus the film expanded in two directions. To study the effect of the different types and content of natural fillers were added into the bio-composite film. Moreover, study the compatibility of the natural fillers and the bio-composite film.

The neat PLA film has the tensile strength at break as 47.48 MPa in MD and TD as 48.61 MPa, %elongation at break in MD and TD were 12.33 and 11.55 respectively. Modulus at break has 681.13 MPa in MD and 639.5 MPa in TD. The addition of natural filler caused all the values of the tensile test were changed that may be affect to changing of the mechanical properties of bio-composite films.

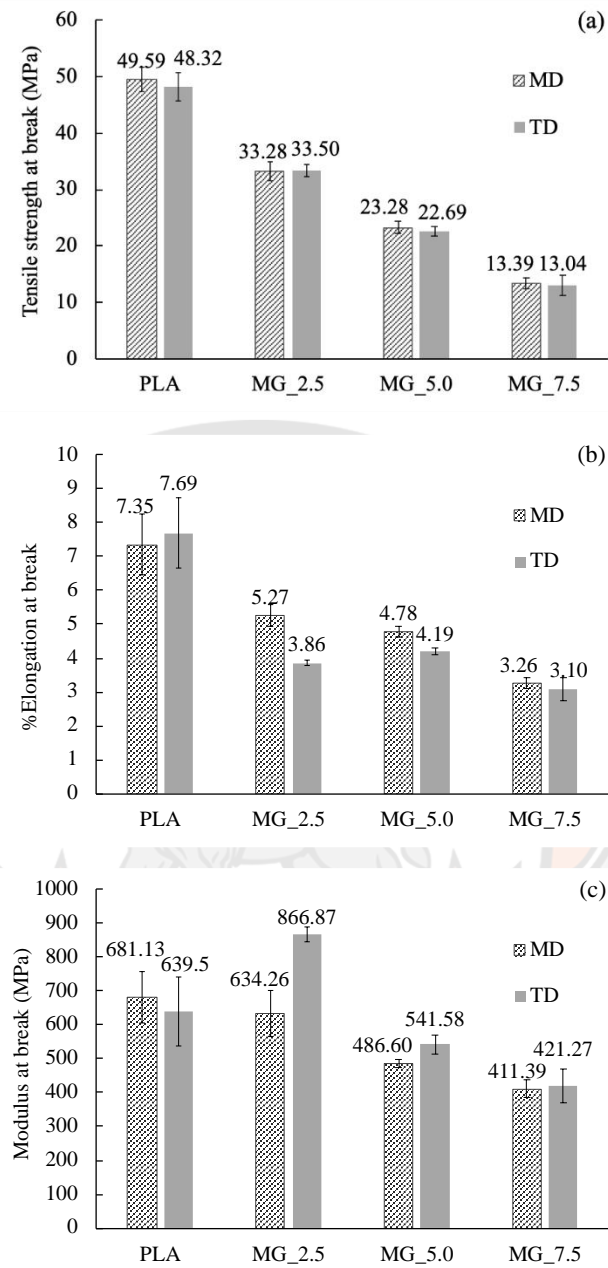
The mechanical properties of the bio-composite film incorporated with EG powder, MG powder and LE powder has shown following Figure 34, 35, and 36, respectively. All the bio-composite films shown the tensile strength was decreased with the highest content of natural fillers in the ranges 9.00-13.00 MPa. Figure 34b, 35b, 36b presents the %elongation at break of the neat PLA film and the bio-composite film. The addition of natural fillers into the bio-composite film causing the strain was decreased which the highest content of natural fillers affects to reduce the %elongation more than 75% when compared with the neat PLA film. The modulus at break of all bio-composite film was lower than the neat PLA film. The modulus at break of the highest content natural fillers adding has approximately 200-400 MPa. The whole natural fillers have the tensile strength, %elongation at break and modulus at break have a slightly different value.

The results are indicated the addition of natural fillers affect the mechanical properties. The reducing of continuous phase of the PLA matrix and the compatibility between the PLA matrix and natural fillers affected from adding fillers into the PLA matrix. The observation of the natural fillers can see in the SEM images in Table 11, 12, 13 are represented the natural fillers are distributed and hinder the continuous phase of PLA. The SEM image shows the interaction between natural particles and the PLA matrix after carried on the tensile testing as the natural particle pulled out from the PLA matrix causing the matrix to have a gap during natural particles and PLA matrix and has some voids on the matrix. These characteristics are contributed affect the mechanical properties that have low value.

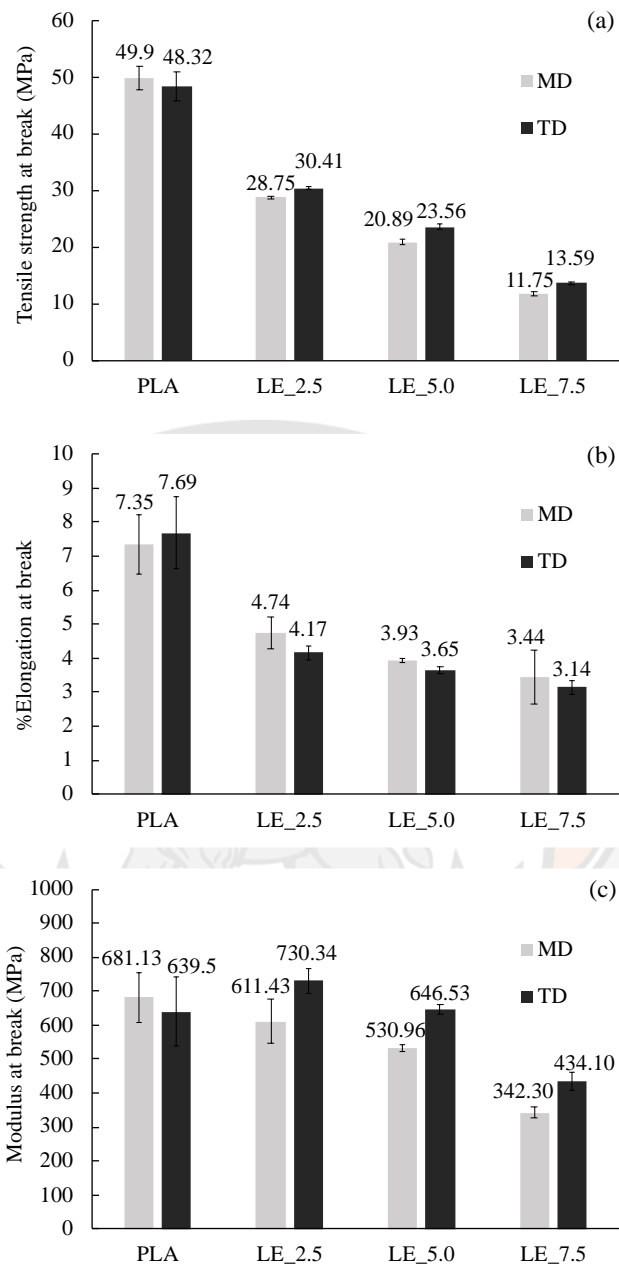
The results can be concluded that all the natural fillers are effect to reducing strength and strain values of bio-composite film that indicated the whole natural fillers are non-reinforcement. For the results of SEM image and melt flow index can be discussed the natural fillers and PLA matrix are immiscible. Then the mechanical properties can investigate all the natural fillers are incompatibility with the PLA matrix. However, the addition of natural filler may be influence reduced the mechanical properties. On the other hand, maybe good effect to the other properties of bio-composite film.



**Figure 34** The mechanical properties of bio-composite film incorporated with EG powder as (a) Tensile strength at break, (b) %Elongation at break, and (c) Modulus at break



**Figure 35** The mechanical properties of bio-composite film incorporated with MG powder as (a) Tensile strength at break, (b) %Elongation at break, and (c) Modulus at break



**Figure 36** The mechanical properties of bio-composite film incorporated with LE powder as (a) Tensile strength at break, (b) %Elongation at break, and (c) Modulus at break

### **The physical properties of the bio-composite film**

The bio-composite films were analyzed the physical properties by water vapor permeability (WVP), color parameter, and UV transmittance. To investigate the different types and content of natural in the bio-composite film that influences the physical properties. The physical properties of bio-composite film were studied then the good natural filler was chosen for improving and developing the bio-composite film.

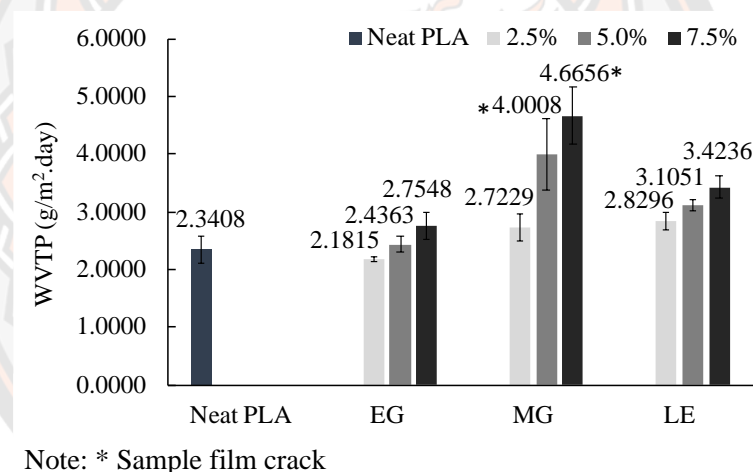
#### 1. Water vapor permeability (WVP)

The bio-composite films were tested the water vapor permeability to determine the effect of different types and content of natural fillers. The water vapor transmission (WVTR) of these films were studied according to ASTM E96-80. WVP is important for the application of mulch film because the mulch film should keep and control the humidity in the soil. To discuss the effect of natural fillers on the water vapor transmission.

Figure 37 show the water vapor transmission (WVTR) of the neat PLA film and the bio-composite film. The neat PLA film has the WVTR as 2.3408  $\text{g/m}^2\cdot\text{day}$  and adding the natural fillers into the PLA matrix affects the WVTR values are changed. The WVTR of bio-composite film with EG powder has value ranges from 2.1000 to 2.8000  $\text{g/m}^2\cdot\text{day}$ . The addition of MG powder has a WVTR of 2.7000-4.7000  $\text{g/m}^2\cdot\text{day}$ . While the bio-composite film incorporated with LE powder has a WVTR ranges on approximately 2.8000-3.5000  $\text{g/m}^2\cdot\text{day}$ . The increasing of natural fillers affects to increasing WVTR.

From the result, the surface and the cross-section morphology are shown in the Table 11-13. The SEM images of the bio-composite film with natural fillers presents the distribution of natural fillers in the PLA matrix which generate many tortuous parts ways in the PLA matrix. The tortuous parts ways influence the vapor diffusion throughout film surface. The increasing of natural fillers affect to the bio-composite film has more particles distribution and generate many filler agglomerates, which reduce the continuous phase in the PLA matrix. Moreover, the agglomerates of natural fillers were poorly distributed affect to the interaction between natural particles and PLA chain that the cross-section surface of the bio-composite film shows a gap and voids. (37, 59) The addition of high content of

natural fillers affects to the natural particles and PLA chain are immiscible. The natural fillers reduce the endangerments of the PLA chain, which affects the orientation of PLA chain. The immiscibility of natural fillers and PLA chain influences the water and vapor can diffusion through out film surface, which affects the WVTR has a high value with the greater of natural filler. Moreover, the increasing of WVTR causes from the chemical components of the natural fillers (cellulose, hemicellulose, and lignin) are hydrophilic compound, therefore these compounds can be absorbed the water and vapor that affects to increasing of WVTR when increase the natural fillers. However, the bio-composite film with MG powder has the highest of WVTR due to the MG film was cracked while in experiment, the sample film covers on the cup was deformation in 3 days thus affects to WVTR has the over rate.



**Figure 37 Water vapor transmission (WVTR) of the neat PLA film and the bio-composite film incorporated with the different types and contents of natural filler**



## 2. Color parameter

The bio-composite films were detected the color parameter (L,  $a^*$ ,  $b^*$ , and  $\Delta E$ ) to analyze the effect of natural fillers on the color shade of the bio-composite film. Which, L value is the lightness of the sample by the positive (+) of L value as the sample has a lightness, whereas the negative (-) of L value is the darkness of the sample. The  $a^*$  value in the color parameter indicated the red/green shade which, positive of  $a^*$  value is red shade and negative of  $a^*$  value is green shade. The  $b^*$  value is the yellow/blue shade that positive of  $b^*$  value assign as yellow shade and negative of  $b^*$  as blue shade. For the  $\Delta E$  shown the difference of color shade between the bio-composite film and the neat PLA film. The adding of natural filler causing the color parameters was changed and influences of the  $\Delta E$  of bio-composite film was changed when compared with the neat PLA film.

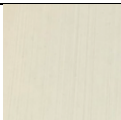


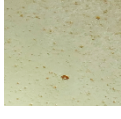

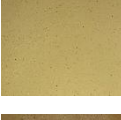

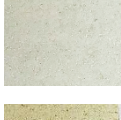


Table 14 is indicated the color parameters of the neat PLA film and the bio-composite film. It was found the neat PLA film has the highest value of lightness as the L,  $a^*$ , and  $b^*$  has  $86.89 \pm 0.34$ ,  $-0.20 \pm 0.00$ , and  $-4.67 \pm 0.07$ , respectively. The addition of natural fillers into the bio-composite film could reduce the lightness of the bio-composite film. Moreover, the effect of natural filler causing the  $a^*$  and  $b^*$  parameters to have increasing value when the content of natural fillers was increased to high content which can observe in Table 14. The effect of parameter changes in L,  $a^*$ , and  $b^*$  causing the  $\Delta E$  of bio-composite film was changed to high value by increasing the content of natural filler.

The bio-composite film incorporated with EG powder has the L value in the ranges 84.00-87.00, ranges of  $a^*$  value as -0.20 to 0.50 and  $b^*$  value between -2.60 to 2.90. The bio-composite film with MG powder has the L,  $a^*$ , and  $b^*$  ranges on as 79.00-84.00, 0.70-2.90, and 3.40-12.30, respectively. For  $\Delta E$  of both bio-composite films include EG film and MG film have the increasing different value of  $\Delta E$  when increasing the content of EG and MG powder which the MG film has different  $\Delta E$  with neat PLA film more than EG film. The EG film and MG film were shown the yellow shade which related to the color shade of ginger because the ginger has the color component as curcuminoids which give the yellow pigment. (44) Moreover, the age of ginger affects the color shade of bio-composite film when observed MG film image in Table 14. The MG film has the dark shade more than EG film because the

harvest time affects the color of ginger which the early immature ginger has the lightness of yellow shade when the harvest time was increased cause the color of ginger was changed to brown shade. (47) Thus, the age of ginger influences the color shade of bio-composite film which the color shade of EG film was the lightness of yellow shade more than the MG film. The addition of LE powder in the bio-composite film affects the color parameters were changed by the L, a\*, and b\* of LE films has ranges on according 77.00-86.00, -0.50-1.40, and 1.80-15.30, respectively. The  $\Delta E$  has a high value more than EG and MG film which the increasing of  $\Delta E$  value with a greater content of LE powder. The LE film was appeared in a green-yellow shade according to the lemongrass leaf color. The lemongrass leaf has chlorophyll A and chlorophyll B that is a green pigment. (49) Furthermore, the content of chlorophyll in the lemongrass leaf has a total content of 1.73 mg/g when let dried the lemongrass leaf powder was found the content of chlorophyll also remain in the particle but found in the low content of chlorophyll as the total content is 0.13 mg/g. (55) So, the color component in the lemongrass affects the color shade of bio-composite film with LE powder appears the green-yellow shade.

The changing of the color parameters affects the performance for UV-transmittance of the bio-composite film. Moreover, the color compound in ginger and lemongrass leaf are influenced to photosynthesis of plant because ginger has curcuminoids as the pigment that can absorb UV-light in a wavelength range of 420-580 nm. (60) The lemongrass leaf has chlorophyll A that can absorb UV-light in the blue-violet region and chlorophyll B can absorb UV-light in red-blue light. (61) Generally, weed and grass must use the UV-light for photosynthesis, (55) but the natural filler has the component for absorbing the light in this range of light thus adding natural filler into the bio-composite film may improve the properties about preventing weed growth and avoid grass on the plot.

**Table 14** The color parameter of the neat PLA film and the bio-composite film

Samples	Color parameters			$\Delta E$	Color
	L	a*	b*		
Neat PLA film	$86.89 \pm 0.34$	$-0.20 \pm 0.00$	$-4.67 \pm 0.07$	Ref.	
EG_2.5	$86.89 \pm 0.17$	$-0.20 \pm 0.00$	$-2.67 \pm 0.28$	2.00	
EG_5.0	$86.64 \pm 0.26$	$-0.02 \pm 0.04$	$-0.81 \pm 0.10$	3.87	
EG_7.5	$84.93 \pm 0.27$	$0.50 \pm 0.15$	$2.86 \pm 0.60$	7.81	
MG_2.5	$84.19 \pm 0.23$	$0.70 \pm 0.05$	$3.45 \pm 0.27$	8.60	
MG_5.0	$81.90 \pm 0.24$	$1.70 \pm 0.05$	$7.80 \pm 0.21$	13.57	
MG_7.5	$79.81 \pm 0.53$	$2.91 \pm 0.16$	$12.25 \pm 0.70$	18.85	
LE_2.5	$85.85 \pm 0.28$	$-0.51 \pm 0.03$	$1.84 \pm 0.31$	6.60	
LE_5.0	$81.67 \pm 0.25$	$0.08 \pm 0.06$	$8.53 \pm 0.34$	14.20	
LE_7.5	$77.39 \pm 0.71$	$1.31 \pm 0.15$	$15.30 \pm 0.94$	22.17	

### 3. UV Transmittance

The bio-composite film was examined the UV-transmittance to determine the effect of natural filler on the UV-light transmittance performance. The bio-composite film was detected in visible regions and can be separated into 3 regions include at 315 nm (UV-B), 400 nm (UV-A), and 700 nm (visible region). %UV-transmittance (%T) of bio-composite film were reported.

Table 15 performs the %UV-transmittance of the neat PLA film and the bio-composite film incorporated with different types of natural fillers and different content for adding. The neat PLA has the highest of %T which the UV-light can transmit through the neat PLA film higher to 90% that cannot protect UV-light. The neat PLA film has transparent properties. When adding the natural fillers into the bio-composite film affects the %T was changed to decreasing the %T. The bio-composite film incorporated with EG powder was expressed the UV-light can transmit through around 11-43% which reduced %T with increasing content of EG powder. Adding MG powder into the bio-composite film has shown the reduction of %UV-transmittance to 6-27%. The reduction of %T of the bio-composite film with MG powder cause by the age of ginger, which the MG powder has a dark shade more than EG powder. The reason also affects the bio-composite film has a less of %T which this result is related to the color parameter of the bio-composite film. The ginger has a compound as curcuminoids that can absorb UV-light in wavelength 420-580 nm. (60) The curcuminoids can help to reduce the transmittance of UV-light through the bio-composite film because the curcuminoids can help to absorb UV-light in this region. Moreover, ginger has more compounds to absorb the UV-light such as gingerol, shogaol, and paradol which is an aromatic compound thus has greater UV-light absorption. (43, 62) Thus, the UV-light also transmittance through the bio-composite film are less content. The incorporation with LE powder of the bio-composite film was performed the light transmittance just 1-20% at the highest content that is the greatest light absorption when compared with whole natural filler for adding. The bio-composite film with LE powder can absorb UV-light more than 80%. That can explain the LE powder that was distributed in the bio-composite film acts as a barrier to UV protection. Because the LE powder has the compounds are chlorophyll A and chlorophyll B that this compound can highly absorb the UV-light in wavelength 450

and 680 nm with chlorophyll A. For chlorophyll B can highly absorb in region 480 nm. (55, 61) Furthermore, the main chemical compound of LE powder as a phenolic compound includes phenols, flavonoids, and alkaloids which these compounds are aromatic compounds that can greatly absorb the UV-light and reduced the UV-light transmittance through to the bio-composite film with LE powder. (52, 62, 63) The addition of natural fillers was bound and distributed in the bio-composite film can improve the UV-light protection because the natural acts as the barrier which can protect the UV-light and reduce the UV-light transmittance. However, the UV-light also can transmittance through to the bio-composite film but the %T was reduced thus the bio-composite film incorporated the natural fillers has the translucent material.

In this work, the bio-composite film was developed and produced for the mulch film. UV-light protection is the main property of mulch film because the planting does not use the mulch film to cover the soil because it is easier to generate the weed and grass growth in the crop. Due to the weed and grass can greatly absorb the UV-light for photosynthesis and well growth. (60) When the UV-light can greatly transmittance influenced the weed can be easier to grow. Causing the farmer must use the herbicide to remove the weed and grass that release the toxic chemical from herbicide to soil and remain in the environment. If the bio-composite film is used to cover the soil that the good effect to the environment which avoid using the herbicide and reduce the toxic of chemical release. Especially, the covering by the bio-composite film can reduce the UV-light transmittance causing can prevent weed growth in the crop as well.

**Table 15 %UV-transmittance of the neat PLA film and the bio-composite film**

Samples	%UV-transmittance		
	at 315 nm (UV-B)	at 400 nm (UV-A)	at 700 nm (Visible region)
Neat PLA film	81.00 ± 1.00	85.33 ± 0.58	90.00 ± 0.00
EG_2.5	35.50 ± 0.00	39.33 ± 0.58	43.00 ± 0.00
EG_5.0	16.00 ± 0.00	19.00 ± 0.00	22.00 ± 0.00
EG_7.5	7.00 ± 0.00	9.00 ± 0.00	11.00 ± 0.00
MG_2.5	17.00 ± 0.00	22.00 ± 0.00	27.00 ± 0.00
MG_5.0	7.00 ± 0.00	10.00 ± 0.00	14.00 ± 0.00
MG_7.5	2.00 ± 0.00	4.00 ± 0.00	6.00 ± 0.00
LE_2.5	27.00 ± 0.00	29.67 ± 0.58	35.00 ± 0.00
LE_5.0	8.00 ± 0.00	9.33 ± 0.58	13.00 ± 0.00
LE_7.5	2.00 ± 0.00	2.00 ± 0.00	4.00 ± 0.00

#### **The biodegradability of the bio-composite film**

The bio-composite films have tested the biodegradability by putting the samples in the soil at 3, 6,9 and 12 months. To study the effect of each natural fillers and different content of natural filler on the biodegradable performance. The recording of sample weight and average the weight change of each samples.

Table 16 presents the sample recovering after during biodegradable in the soil. It was found the overtime causing all the samples to have a slight color changing. Almost all the bio-composite film remained the shape except the bio-composite film incorporated with MG powder. The percentage weight loss of the bio-composite film after biodegradation in 3, 6, 9 and 12 months was shown in Figure 38. The result showed the tendency for weight loss of the bio-composite films was increased when increasing biodegradation times. The neat PLA film rather less degradable which degrades just 1.09 % weight loss after burying in the soil for 12 months. The addition of natural can help improve the biodegradability of the bio-composite film. The bio-composite film with natural filler can degrade more than neat PLA film when observe

in Figure 38 was shown percentage of weight loss. It was found the bio-composite film incorporated with whole natural filler have a high percentage weight loss more than neat PLA film.

The bio-composite film incorporated with EG powder after 6 months has enhancement of the percentage weight loss and increasing content of EG powder influences the percentage weight loss more increased. The highest content of EG powder (7.5% w/w) at 6 months has percentage weight loss at 2.85% overtimes after 6 months has increased percentage weight loss to 6.79% and after burying in soil 12 months were found the bio-composite film with EG powder can degrade to 7.86%. The bio-composite film adding with MG powder has a tendency like the bio-composite film with EG powder. However, the addition of MG powder has the higher degradation more than EG powder that the sample after 6 months buries in soil and the bio-composite film has fast degradable when the sample was fragmented to small size that can see the fragment samples in Table 16. The weight of sample was recovered after 6 months that shown the increasing of percentage weight loss more to 46.34% from 2.03% at 3 months. The increasing time for burying in soil that can the greatest degrade as showing after 12 months and the percentage weight loss increases to 63.40%. The comparison of the bio-composite film with MG powder has the percentage of weight loss more than incorporated with EG powder that it maybe effect from MG particles and the PLA matrix were separated together and have a less continuous phase which related to the morphology of the bio-composite film can see in SEM image. This immiscibility of the bio-composite film and MG powder maybe affect the properties of film that easier to cracking. Another reason is related WVP of the bio-composite film with MG powder has a high value of WVTR due to the morphology of bio-composite film has gap and hole between MG particle and PLA matrix. The relation during the bio-composite film and MG powder causing the sample is easier to fragmentation and disintegration into small pieces. Moreover, the bio-composite film incorporated with LE powder that presented the same trend for the percentage of weight loss with the bio-composite film with both ages of ginger. The increment of burying in the soil causing the sample has more degradable which performed more enhancement of the percentage of weight loss. In addition, the high content of LE powder can help to degradable. The samples of the bio-composite film

with the highest content of LE powder after buries in soil 9 months have fast degradable which has the percentage of weight loss increase to 14.59%. After 12 months, the bio-composite film with LE powder can high up degradable as 16.58% of weight loss.

Generally, PLA is a biodegradable polymer that can be degraded via the hydrolysis process. The suitable condition includes the moisture, microorganism, and content of oxygen was influenced the degradation of PLA. Which, PLA will degrade to water, carbon dioxide, and another compound with non-toxic chemicals. (33) The addition of natural fillers into the PLA matrix has influenced the degradability of neat PLA film. The natural fillers with the PLA can support fast decompose properties that can observe in the tendency for the percentage of weight loss in Figure 48. The high up degradable of the bio-composite film when adding the content of natural fillers because the natural fillers maybe hinder the continuous phase of PLA matrix causing the strength of bio-composite film was reduced. The observation in the mechanical properties of the bio-composite film shown the tensile strength and %elongation of a whole bio-composite films was decreased thus, effect to the bio-composite film has a brittle and easier to fragmentation. Moreover, in this work performed testing by burying in the soil due to the moisture and microorganism thus effect to the degradability that sample can be fragment to small size. From the tendency, if increasing time of degradable causing the sample maybe can degradable high up and help to reduce to the small size, moreover, the last maybe degrade to water and carbon dioxide, respectively.



**Table 16 The bio-composite film sample recovered after biodegradable in soil**



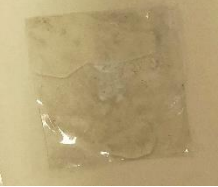













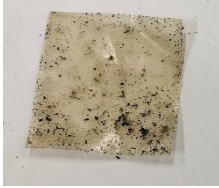







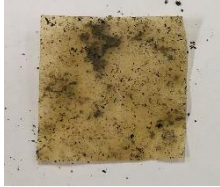















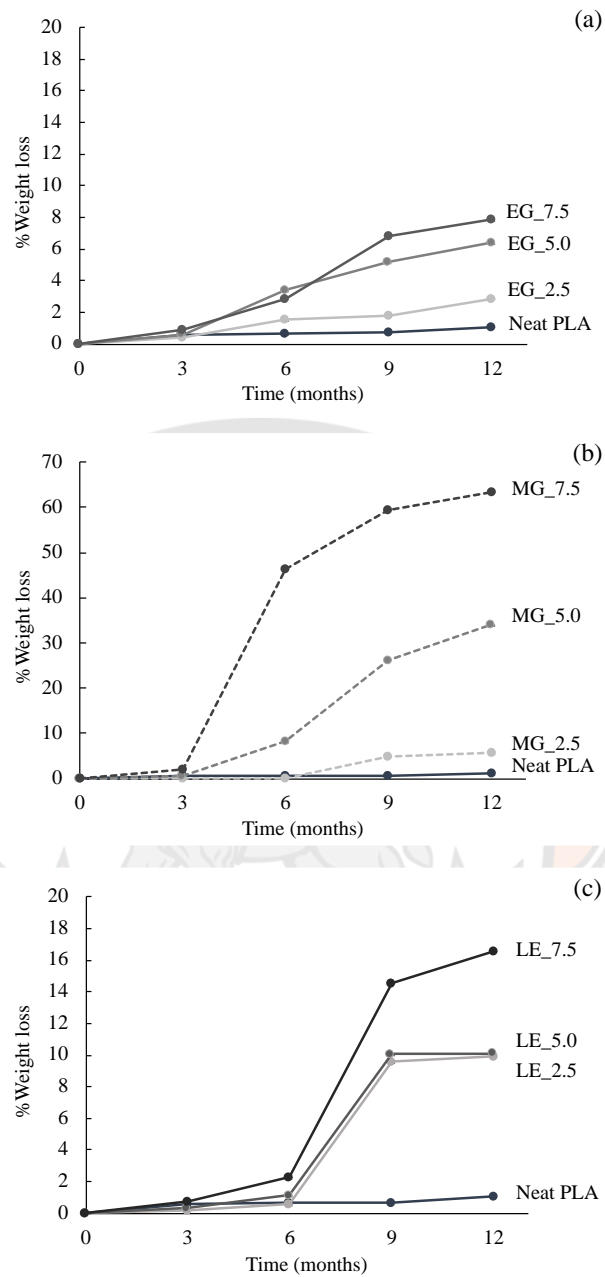
Sample	Biodegradation times (months)			
	3	6	9	12
Neat PLA film				
EG_2.5				
EG_5.0				
EG_7.5				
MG_2.5				
MG_5.0				

Table 16 (Cont.)

Sample	Biodegradation times (months)			
	3	6	9	12
MG_7.5				
LE_2.5				
LE_5.0				
LE_7.5				



**Figure 38 Biodegradability of the neat PLA film and the bio-composite film incorporated with (a) MG powder, (b) EG powder, and (c) LE powder**

### **The effect of polybutylene succinate (PBS) in the bio-polymer blend base on PLA matrix**

The purpose of this section was studied the effect of polybutylene succinate (PBS) in bio-polymer on the chemical properties, physical properties, mechanical properties, and thermal properties. The variation of PBS was presented 0, 10, and 20 %w/w. The results in this section can be separated into 2 kinds including bio-polymer blend compound and bio-polymer blend film. Due to there are many kinds of bio-polymer blends. Abbreviations were used to represent in each sample. For example, poly(lactic acid) or PLA will use abbreviation ‘L’ letter followed by the content of PLA as a number. The “B” letter will be polybutylene succinate or PBS following the percentage of PBS content as number. All abbreviation of the samples were shown as following in Table 17.

**Table 17 The acronym of bio-polymer blend compound and film**

<b>Content of PBS (%w/w)</b>	<b>Acronym</b>
0	PLA
10.0	L90_B10
20.0	L80_B20

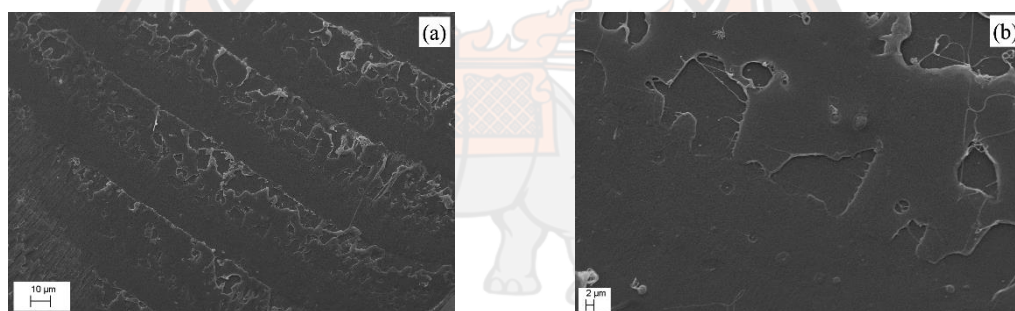
#### **1. Bio-polymer blend compound**

The sample of the bio-polymer blend compound was performed by a twin-screw extruder. The miscibility of PBS in the bio-polymer blend based on PLA was studied by using scanning electron microscopy (SEM). Moreover, the melt flow index (MFI) technique of the bio-polymer blend compound was studied.

### Morphology of bio-polymer blend compound

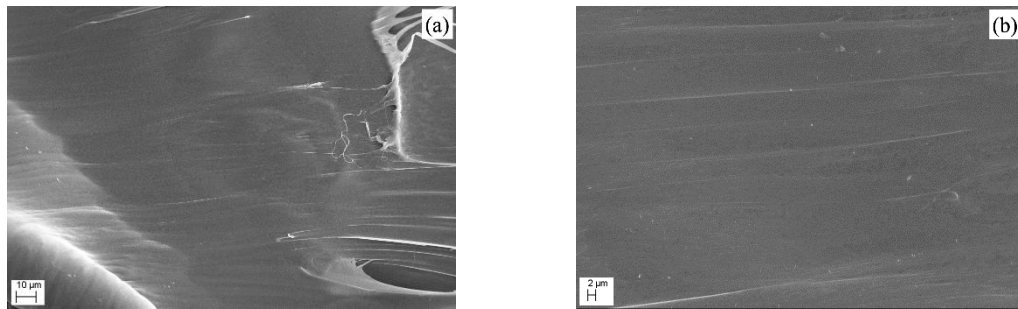
Morphology of the bio-polymer blend based on PLA was studied in the form of cross-section. Bio-polymer blend compound from pellet cutting was prepared by the twin-screw extruder. Moreover, variation of content of PBS affects the miscibility between PLA and PBS were discussed. The bio-polymer blend compound was examined by scanning electron microscopy (SEM) at magnification 200X and 1000X.

The cross-section morphology of the neat PLA compounds and bio-polymer blend compounds from twin-screw extruder present in Figure 39-41. The pellet cross-section compound of the neat PLA shows the clear surface, non-appear voids, and have homogeneous phase. The results were shown in Figure 39a-b.



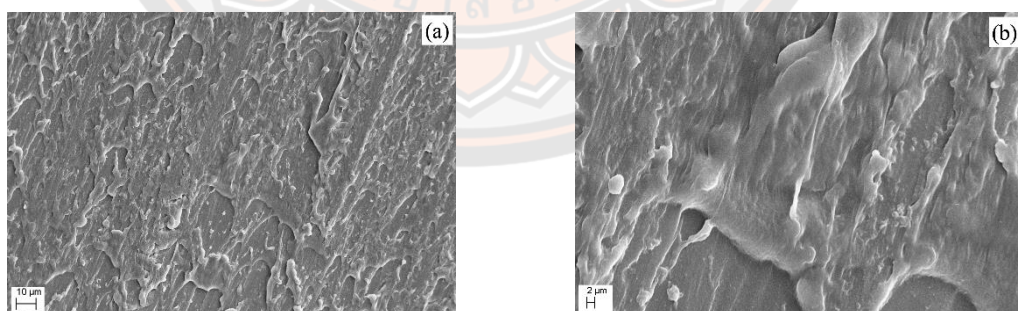
**Figure 39 cross-section morphology of the neat PLA compound at magnification (a) 200X, and (b) 1000X**

The addition of PBS into the PLA based also affects the properties of the bio-polymer blend. The bio-polymer blend with 10% w/w of PBS shows the cross-section morphology in Figure 40. Cross-section morphology has clear surface, non-voids, and the polymer is stretches. The addition of 10% w/w of PBS shows the cross-section morphology has surface miscibility.



**Figure 40** Cross-section morphology of the bio-polymer blend based on PLA with 10%w/w of PBS content at magnification (a) 200X, and (b) 1000X

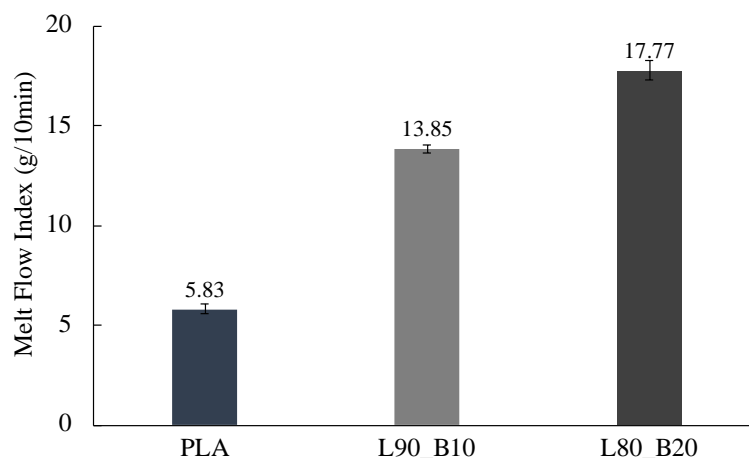
On the other hand, when adding content of PBS to 20%w/w shows the cross-section morphology has roughness that can be observed in Figure 41. Moreover, the phases of the bio-polymer blend compound were separated from the continuous phase. The SEM image of the bio-polymer blend can be discussed the adding of 10% of PBS into the PLA base influences the bio-polymer blend has miscibility. While the content of PBS adding more than 10% affects to the bio-polymer blend has the immiscibility. These characteristics of bio-polymer blend are influenced the melt flow index and mechanical properties of the bio-polymer blend film. (40)



**Figure 41** Cross-section morphology of the bio-polymer blend based on PLA with 20%w/w of PBS content at magnification (a) 200X, and (b) 1000X

### **The effect of PBS content on melt flow index (MFI)**

Melt flow index (MFI) of the bio-polymer blend compound was studied follow ASTM D1238E. The influence of the content of PBS of melt flow index was studied and results were changed and shown in Figure 42. The results show the MFI of the neat PLA compound was 5.83 g/10min while the addition of PBS content to PLA based may caused the MFI of bio-polymer blend was changed to increase values. However, addition of the content of PBS with 10%w/w causing the MFI value more than the neat PLA compound, which MFI was increased to 13.85 g/10min. The increasing content of PBS causing the greater MFI value which the MFI of bio-polymer blend with 20%w/w of PBS was 17.77 g/10min. The increasing of MFI when adding more content of PBS due to PBS is an aliphatic polyester polymer which structure consists of succinic acid and 1,4-butanediol that the chain of PBS has number of carbons more than PLA chain because PBS has high flexibility more than PLA. (7) When increasing the content of PBS for adding into the PLA based that the PBS could improve the chain mobility of PLA and the bio-polymer blend can be easier to flow causing the MFI was increasing value. However, when adding more content of PBS to 20%w/w will affect the bio-polymer blend that has high value of MFI. It will indicate the PLA and high content of PBS was immiscible. Immiscibility of PLA and 20%w/w of PBS content in the result can be observed by SEM image in Figure 41. The results show that immiscibility of the PLA and PBS blend has roughness and phase separate when adding high content of PBS in Figure 43.



**Figure 42 Melt flow index of the bio-polymer blend based on PLA with varies content of PBS**

## **2. Bio-polymer blend film**

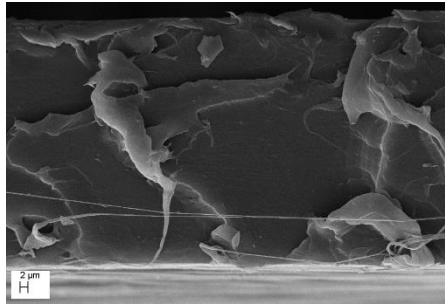
The bio-polymer blend films were analyzed the physical, mechanical, and thermal properties. Different content of PBS in the bio-polymer blend film was investigated influences miscibility of the polymer blend. Moreover, optimum ratio of PLA and PBS was chosen for developing the bio-composite film.

### **Morphology of bio-polymer blend film**

Morphology in form of cross-section of the bio-polymer blend film after tensile testing was studied. The investigation of the various content of PBS affects to the morphology and discussion about the miscibility between PLA and PBS. The samples were analyzed by scanning electron microscopy (SEM) at magnification 1000X.

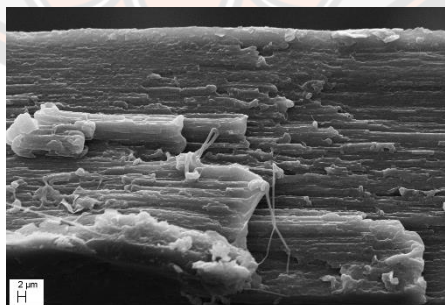
Figure 43 was performed the and cross-section morphology of the neat PLA film. The film sample was examined the mechanical properties by tensile testing to investigate the characteristic after carried on tension force. The cross-section of the neat PLA film is slightly stretched after carried on tensile testing. The cross-section morphology shows a homogeneous phase.





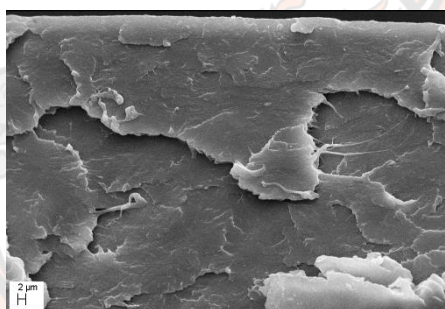
**Figure 43 The cross-section morphology of the neat PLA film at magnification 1000X**

Adding the content of PBS affects the cross-section morphology of the biopolymer blend film is slightly changed. Figure 44 presents the cross-section morphology of the PLA blend with 10% w/w PBS (L90\_B10). The cross-section morphology is slightly stretched after received tension force. The phase of L90\_B10 after tensile testing has non-separated between PLA and PBS and the cross-section morphology has a smooth surface but less smooth than the cross-section of the neat PLA film. The cross-section morphology of the L90\_B10 compound by the cutting pellet from the twin-screw extruder was shown in Figure 40. So, the PLA blend with 10% w/w has rather miscibility with influences the mechanical properties of the biopolymer film.



**Figure 44 The cross-section morphology of the L90\_B10 film at magnification 1000X**

The increasing content of PBS more than 10%w/w into the PLA base the morphology and the cross-section morphology of the bio-polymer blend film. Figure 45 is the PLA blend with 20%w/w of PBS (L80\_B20) show the cross-section morphology. The cross-section morphology of the L80\_B20 presents separated phase after carried on tensile testing. The SEM image according to the characteristic of the cross-section of the L80\_B20 compound has the phase-separated between PLA and PBS. The bio-polymer blend between PLA and 20%w/w of PBS has rather an immiscibility. The addition of 10%w/w PBS has a good surface morphology more than adding at 20%w/w of PBS content. So, the PLA blend with 10%w/w has more miscible and probably can be improved and developed the mechanical properties of neat PLA film.



**Figure 45 The cross-section morphology of the L80\_B20 film at magnification 1000X**

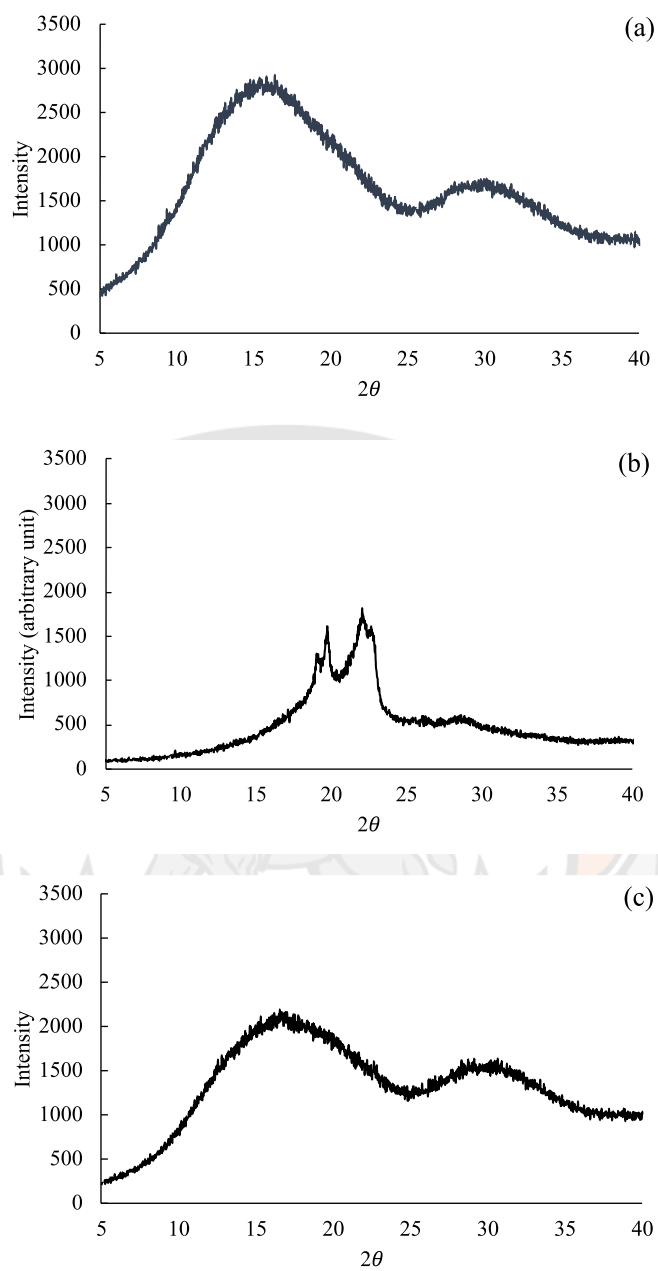
## **2. Chemical properties of bio-polymer blend film**

Chemical properties of bio-polymer blend film were performed by X-ray diffraction (XRD). Adding of PBS was varied content from 0, 10, and 20%w/w were studied. The XRD results show the effect of increasing content of PBS causing the intensity of crystallinity of bio-polymer blend.

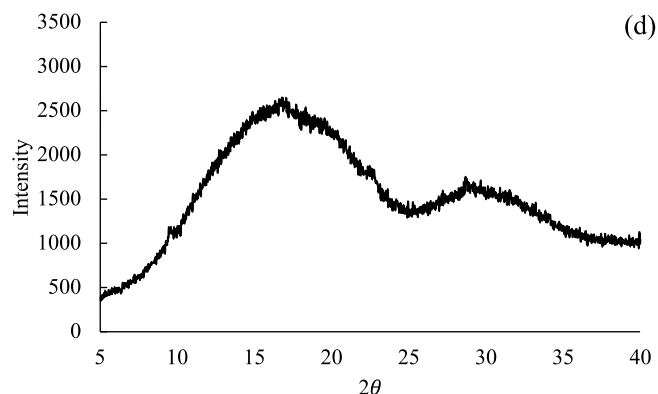
Figure 46 shows the XRD pattern of the neat PLA film and the bio-polymer blend film. The neat PLA film was indicated the XRD peak has 2 main peaks consists of at  $2\theta = 16.36^\circ$  and  $2\theta = 30.78^\circ$ . At  $16.36^\circ$  is the highest intensity approximately 2900 and at  $20.78^\circ$  has an intensity of around 1700 that was shown in Figure 46a. The neat PBS film appear 2 mains of the XRD peak consists of  $2\theta =$

19.77° and  $2\theta = 22.05^\circ$ . The diffractogram of PBS has sharp peak more than PLA because the structure of PBS has the crystalline more than PLA that shown in Figure 46b. The addition of PBS into the PLA base affects the intensity of  $2\theta$  was changed. Figure 46c presents the diffractogram of the PLA blend with 10%w/w of PBS was expressed 2 main peaks consist of approximately 16.52° and 30.42°. The intensity of each peaks also less than the neat PLA film. The amorphous of the bio-polymer blend was reduced due to the structure of PBS has crystalline around 33-45%. Thus, the bio-polymer blend film with 10% PBS has promising increase of the crystalline, which relate to the DSC of L90\_B10. However, the increasing content of PBS to adding onto PLA base in Figure 46d shows the PLA blend with 20%w/w of PBS. It was found the XRD pattern also has 2 main peaks include 16.85° and 29.65°, which has high intensity more than the L90\_B10 film. Moreover, the peak from diffractogram has a large broad band more than the neat PLA film and L90\_B10.

The result was explained the addition of over content of PBS into the PLA based causing reduced crystallinity of the sample that may be influenced the mechanical properties of the bio-polymer blend film. However, the diffractograms shows the main peak of all film samples have the broad band, which investigated the neat PLA film, and all-bio-polymer blend film characteristic are amorphous polymer. Because the adding of PBS content into PLA based was caused to reduce the continuous phase of PLA that shown in SEM image. Moreover, the PBS chain may be hidden the PLA chain also reduce the crystallinity of the bio-polymer blend. This characteristic affects the mechanical properties and thermal properties of the bio-polymer blend film.



**Figure 46 XRD pattern of (a) neat PLA film, (b) PBS film, (c) L90\_B10, and (d) L80\_B20**



**Figure 46 (Cont.)**

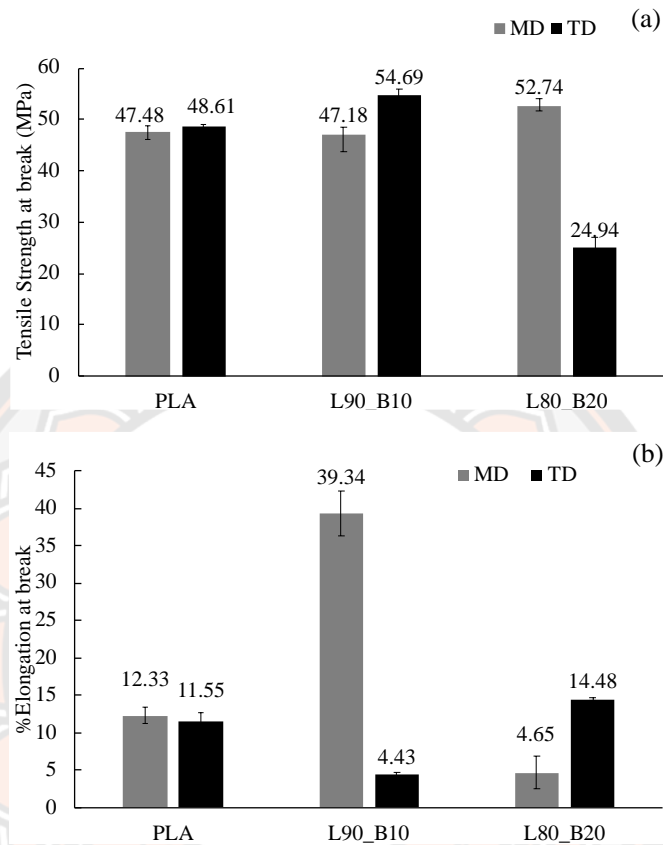
### **Mechanical properties of bio-polymer blend film**

The mechanical properties of neat PLA film and the bio-polymer blend film were examined via tensile testing. The sample characterization was tested in 2 directions consist of mechanical direction (MD) and transverse direction (TD). The result was discussed on the tensile strength at break, %elongation at break, and modulus at break. The effect of the PBS content adding into PLA based was investigated via the mechanical properties. Furthermore, compatibility between PLA and PBS was also discussed via the mechanical properties.

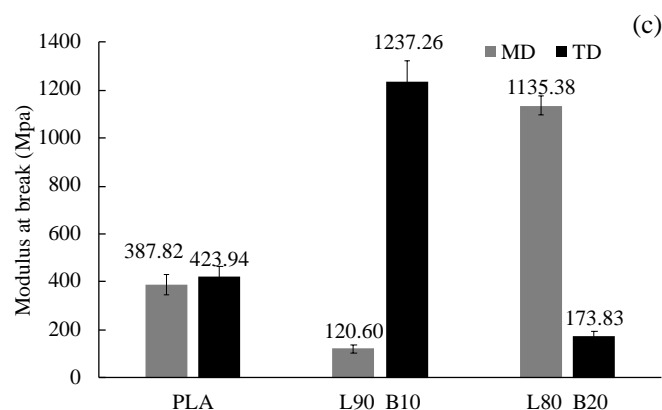
Figure 47 represents the mechanical properties of the neat PLA film and the bio-polymer blend. It was found the tensile strength at break of the bio-polymer blend film was increased more than the neat PLA film, which neat PLA film shows the tensile strength at break as 47.48 and 48.61 MPa in MD and TD, respectively. When adding the content of PBS with 10% w/w affect increasing the tensile strength at break to 47.18 MPa in MD and 54.69 MPa in TD. However, when increasing the content of PBS to 20%w/w causing the tensile strength at break was more decreased in TD. This case may cause the high content of PBS affects reducing the compatibility of the bio-polymer blend during PLA and PBS. According to the melt flow index (MFI) of the bio-polymer blend, adding more over the content of PBS will affect the bio-polymer blend and easier to melt flow because the PLA chain was hidden from the PBS chain. Another agreement is the SEM image was shown the immiscibility morphology when adding over the content of PBS can observe in Figure 41. The observation with the

cross-section morphology of L80\_B20 was found the sample after carried on tension force has the phase-separated can see in Figure 45. The high content of PBS may be caused the interaction between PLA and PBS particle that has poor interaction and reduce the tensile strength at break of L80\_B20. %Elongation at break of the neat PLA film and bio-polymer blend film was shown in Figure 47b. The additional content of PBS with 10%w/w affects the increase of %elongation at break in MD. While in the TD was found %elongation at break was reduced because the specimen during tensile testing has cracked without stretch behavior. Which this case has agreed with the PLA properties because the PLA has hard and brittle. Thus, the crack behavior thus does not stretch. (7) The increasing content of PBS to 20%w/w shows the low %elongation at break in MD because the PBS particles reduce the continuous phase and causing the phase has separate can observe in Figure 45. So, the effect of the high content of PBS has reduced the chain mobility of PLA that influenced the specimen was reduced stretch and show high the tensile strength at break. On the other hand, the TD shows a high %elongation at break that according to the properties of PBS is flexible. The high of %elongation at break in TD may be the specimen after carried on the tension force has rearrangements of PBS chain causing the bio-polymer blend film has increased the stretch. The specimen presents ductile properties thus it has less tensile strength, which the results are related follow with the principle of the bio-polymer blend. (64) This result relates to the XRD result that the addition of PBS causing some direction has increased the %elongation. The observation of XRD pattern in Figure 46 was found the adding more content of PBS causing the bio-polymer blend to have a broader band that meaning the bio-polymer blend film has more amorphous. The modulus at break shown in Figure 47c, which shows the neat PLA film has a high modulus more than the bio-polymer blend film when compare with L90\_B10 in MD and L80\_B20 in TD. The high modulus at break of the neat PLA follows the hard and brittle properties. While the adding of PBS content causing the modulus at break has a lower value than the neat PLA film in MD at 10%w/w of PBS and decrease in TD at 20%w/w of PBS. Thus, the addition of PBS content can help to improve the flexibility and the stretching of the bio-polymer blend film. In addition, the adding content of PBS at 10%w/w is the appropriate ratio between PLA and PBS. Which the bio-polymer blend film between PLA and PBS of 10%w/w can

compatible when adding more than 10%w/w causing the bio-polymer blend film to come to incompatibility.



**Figure 47** The mechanical properties of the neat PLA film and the bio-polymer blend film as (a) tensile strength at break, (b) %elongation at break, and (c) modulus at break



**Figure 47 (Cont.)**

### **Thermal properties of bio-polymer blend film**

The neat PLA film, neat PBS film, and the bio-polymer blend film were characterized thermal properties by differential scanning calorimeter (DSC). In the first heat of the experiment is to clear the thermal history of the polymer. The glass temperature ( $T_g$ ), cold crystallization temperature ( $T_{cc}$ ), melting temperature ( $T_m$ ), enthalpy of melting ( $\Delta H_m$ ), and the degree of crystallinity ( $\%X_c$ ) were studied in the second heat to discuss the miscibility of the bio-polymer blend.

Table 18 shows the data of DSC analysis of the neat PLA film, neat PBS film, and bio-polymer blend film. The neat PLA film show  $T_g$  on 57.04 °C. The neat PBS film was not appeared  $T_g$  due to PBS has the low  $T_g$  with around -30 °C but this work carried heating from 25-200 °C. The addition of PBS content in the PLA base was found the  $T_g$  of the biopolymer film was changed.  $T_g$  of L90\_B10 was shifted to a lower temperature as 48.07 °C.  $T_g$  of L80\_B10 has a visible separated to 2 peaks which  $T_g$  appears around 48.22 °C and 71.19 °C.

$T_{cc}$  of the neat PLA film shows 103.18 °C. The additional content of PBS causing  $T_{cc}$  of the bio-polymer blend was changed to a lower temperature. While neat PBS was not appeared  $T_{cc}$  in the thermogram. The adding content of PBS as 10%w/w causing  $T_{cc}$  born around 92.34 °C. The increasing content of PBS to 20%w/w shows  $T_{cc}$  around 96.99 °C. Cold crystallization temperature ( $T_{cc}$ ) after adding content of PBS into PLA based was shifted. The addition of PBS influences was promoted the crystalline generation. Addition of PBS into the PLA base affects the polymer to



blend it easier to generate the crystalline thus can discuss PBS act as the nucleating agent. (65)

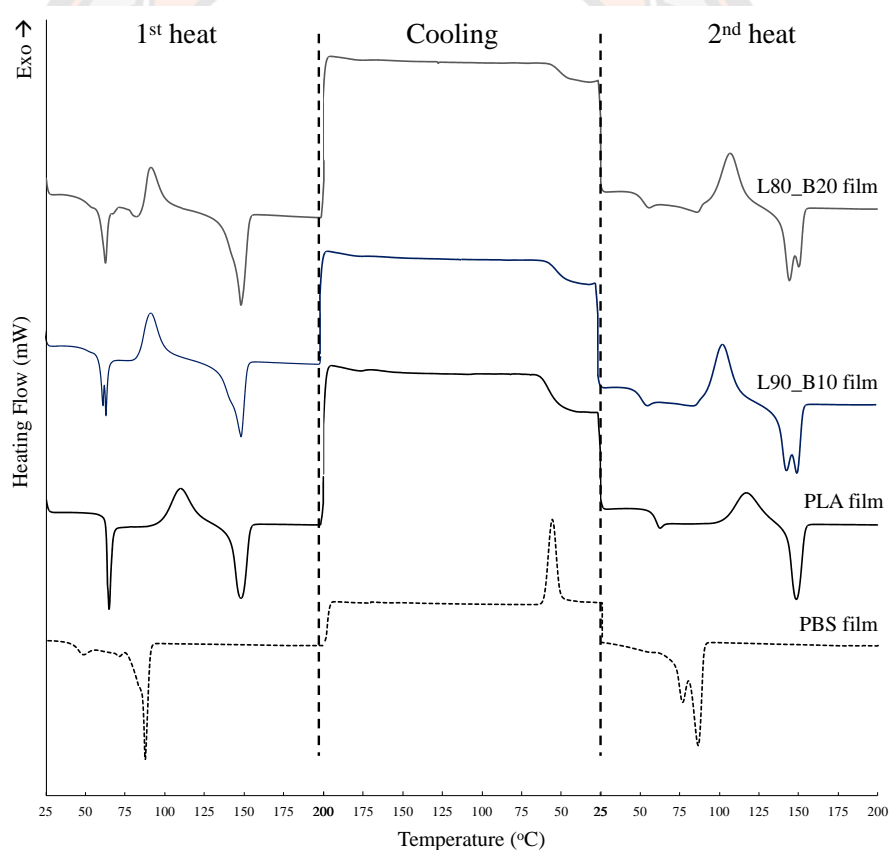
While  $T_m$  of the neat PLA promotes approximately 142.82 °C and neat PBS shows around 72.84 °C. Increasing content of PBS for adding into PLA base affects the melting temperature of the bio-polymer blend.  $T_m$  of adding PBS at 10%w/w was shifted to 141.78 °C and adding of 20%w/w causing  $T_m$  was shifted to a lower temperature. Moreover, the addition of PBS affects  $T_m$ , which changed and appeared two distinct peaks. The results were shown in Figure 48c. From  $T_g$  and  $T_m$  of the bio-polymer blend can be discussed whole of the bio-polymer blend has a spilled peak of  $T_g$  and  $T_m$  peak. Increasing content of PBS for adding influence  $T_g$  of L80\_B20 was obviously separated into 2 peaks. In the summary, the addition of PBS more than 10%w/w causing the bio-polymer blend between PLA and PBS to come to immiscibility. This result related to the melt flow index and the morphology of the L80\_B20.

The calculation of the crystallinity ( $\%X_c$ ) of the neat PLA film has 22.95% and the neat PBS has 10.05%. The additional content of PBS has affected the crystallinity of the bio-polymer blend. The addition of PBS as 10%w/w and 20%w/w causing the increase of  $\%X_c$  that increase to 32.37% and 32.92%, respectively. The results relate to the XRD result, which adds contents of PBS affects to reducing the intensity of amorphous peak.

So, all the results in this work have been interested to choose the good ratio of PLA and PBS is 90: 10 due to this ratio has a miscible and good mechanical property. Addition of PBS at 10%w/w can be improved the strength and the flexibility of the bio-polymer blend.

**Table 18 Data of DSC analysis of the neat PLA film, the neat PBS film, and the bio-polymer blend film**

Sample	2 <sup>nd</sup> heat				
	T <sub>g</sub>	T <sub>cc</sub>	T <sub>m</sub>	ΔH <sub>m</sub>	%X <sub>c</sub>
PLA	57.04	103.18	142.82	21.34	22.95
PBS	N/A	N/A	72.84	21.11	10.05
L90_B10	48.07	92.34	141.78	27.09	32.37
L80_B20	48.22	96.99	139.05	24.49	32.92



**Figure 48 DSC thermograms of the neat PLA film, the neat PBS film, and the bio-polymer blend film**

### **The effect of polybutylene succinate (PBS) and natural filler as lemongrass leaf powder (LE) in the bio-composite base on PLA matrix**

From above section was studied the properties of each natural filler and improving the properties of PLA by adding content of PBS. All of the results can be discussed and chosen the type of natural filler as lemongrass leaf powder, which shows the miscible with the PLA matrix. Moreover, the appropriate ratio between PLA and PBS is 10%w/w of PBS has miscibility and compatibility with the PLA matrix. So, in this part has been interested to choose the bio-polymer blend between PLA and PBS as 90: 10 and choosing the lemongrass leaf powder as a natural filler to adding into the bio-composite.

This section has been interested to produce and develop the mulch film from bio-composite film based on the PLA matrix. The properties of the PLA to flexible by adding content of PBS at 10%w/w was improved. Moreover, the lemongrass leaf powder as natural filler was added to improve the color and UV-light transmittance of the bio-composite film. The effect of PBS content at 10%w/w and various natural filler content as 0, 2.5, 5.0, 7.5, and 10.0%w/w were studied. This work was assumed the PBS and lemongrass leaf powder act as fillers for adding into the polymer matrix as PLA. In summary, chemical properties, mechanical properties, physical properties, thermal properties, biodegradability, aging test, and field test were examined.

The abbreviation of this part using the first letter is “B10” represent the content of PBS at 10%w/w. Another letter is “LE” represented lemongrass leaf powder as natural filler which follows the content of natural filler as a number. All abbreviation of samples were shown in Table 19.

**Table 19 The acronym of the bio-composite with PBS and various content of lemongrass leaf powder**

Content of PBS (%w/w)	Content of LE (%w/w)	Acronym
	0.0	L90_B10
	2.5	B10_LE2.5
10.0	5.0	B10_LE5.0
	7.5	B10_LE7.5
	10.0	B10_LE10.0

### **1. Bio-composite compound from PLA, PBS, and LE powder**

The pellets bio-composite compound from a twin-screw extruder was used to study. The morphology surface was analyzed by scanning electron microscopy (SEM) to observe the preliminary miscibility of the bio-composite. The melt flow index (MFI) of the bio-composite compound was investigated.

#### **Morphology of bio-composite compound**

Attributed of the distribution of the PBS particles and the LE powder in the PLA matrix from the cross-section pellet from a twin-screw extruder were studied. Moreover, interaction between PLA matrix and fillers as PBS, and LE powder to considers the preliminary miscibility of the bio-composite film were discussed. The cross-section pellet was characterized by using scanning electron microscopy (SEM) at magnification 200X for observing the overall cross-section morphology and PBS particles and LE powder distribution. At magnification 1000X for discuss the interaction between PLA, PBS, and LE powder.

Table 20 represents the cross-section morphology of the neat PLA compound and each bio-composite compound at magnification 200X and 1000X. The neat PLA compound has a continuous phase and clear surface, which looks in Table 20a-b. The addition of PBS into the PLA matrix that appears the surface has a continuous phase, clear surface, and non-voids. Addition of LE powder into the matrix affects the cross-section morphology of the bio-composite was changed to

roughness. All of the cross-section morphology of the bio-composite compounds expresses the LE particles bind in the matrix and the matrix has a single phase. The increasing content of LE powder affects to the morphology have more roughness. All of bio-composite compounds show the LE particles are distributed in the matrix. The increasing content of LE powder influences the LE particles are generated the agglomerate in the matrix. The addition of the LE powder of more than 5%w/w presents the hole and gap between the matrix and LE particles. The highest content of LE powder was observed in Table 20k-l, some of the LE particles pull out from the matrix and present a gap between LE particles and polymer matrix. Generally, the lemongrass leaf has the content of cellulose 39.5%, hemicellulose 22.6%, and lignin 28.5% that whole compounds are the hydrophilic compound. (47, 56, 57) High of hydrophilic compound cause to easily generate the agglomerate in the matrix. The continuous phase is reduced by the LE agglomerates. Moreover, the different shapes and variation of sizes affects the distribution of polymer matrix. The SEM images in Table 20 show the rod shape and long particle of LE powder that distribute in the polymer matrix and the rod particle pulled out of the matrix in orientation following the machine. The rod shape binds in the polymer matrix that generated the gap between particle and polymer matrix.

In the summary, addition of more content of LE powder as more than 5%w/w causing the bio-composite between matrix and LE particle come to immiscibility. The over LE powder content affects the distribution in the matrix has many LE agglomerates. While addition of PBS has not shown the PBS particle in the matrix. PBS particles bound in the PLA matrix and the phase non-separated together.

**Table 20** Cross-section morphology of the neat PLA compound and the bio-composite compound

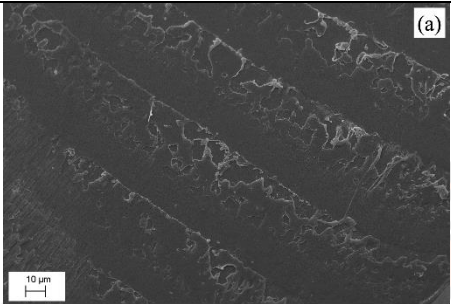
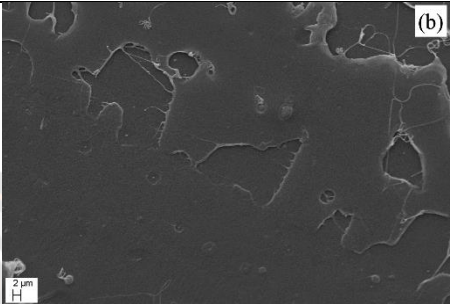

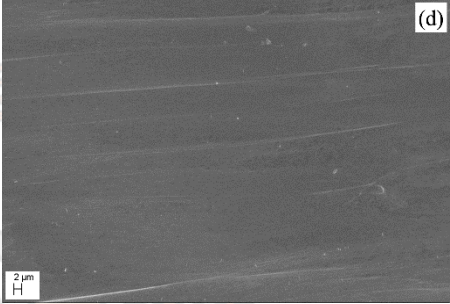
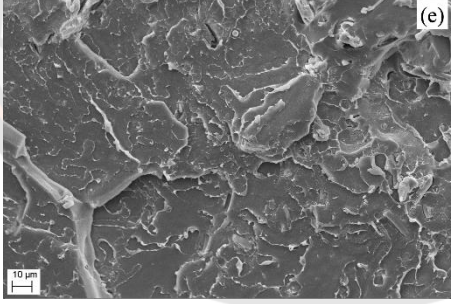
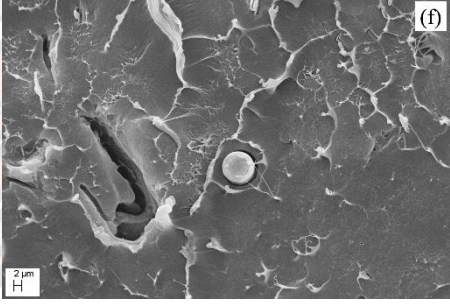
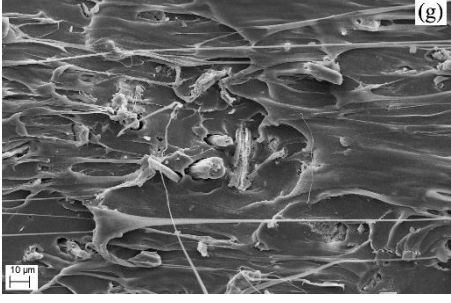
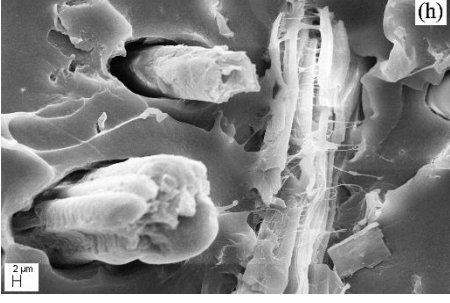
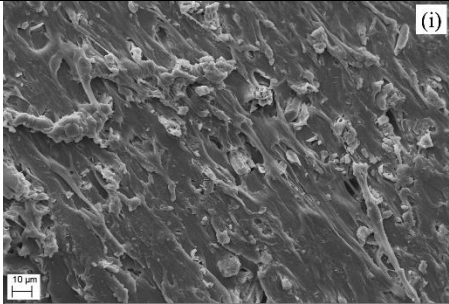
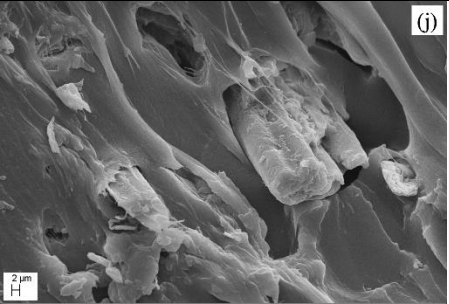
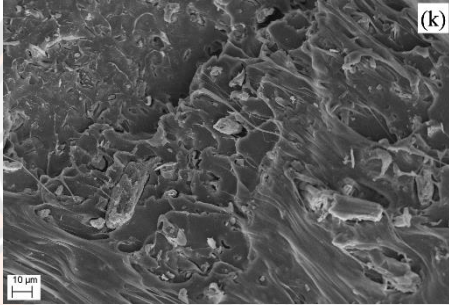
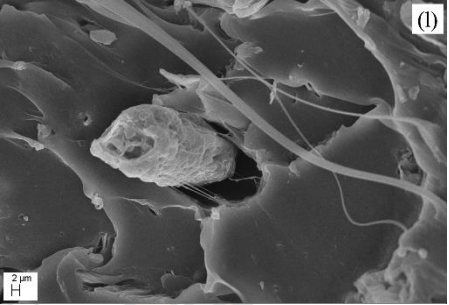
Sample	Magnification	
	200X	1000X
Neat PLA		
L90_B10		
B10_LE2.5		
B10_LE5.0		

Table 20 (Cont.)

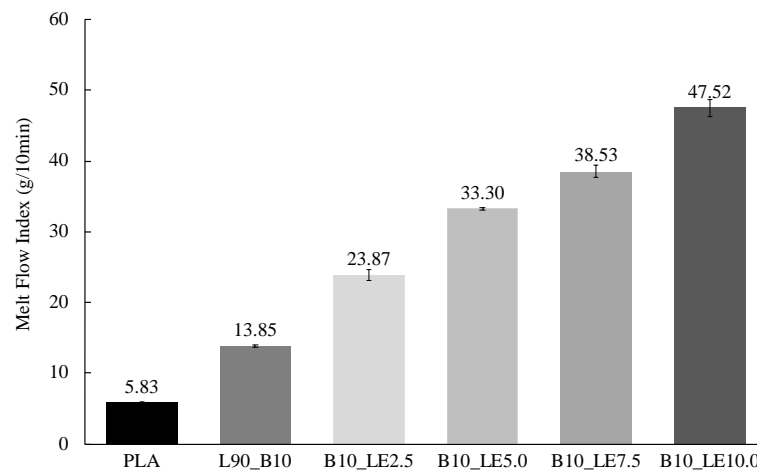
Sample	Magnification	
	200X	1000X
B10_LE7.5		
B10_LE10.0		

### Melt flow index (MFI) of the bio-composite compound based on PLA matrix incorporated PBS and LE powder

The influence of the PBS and the various content of LE powder also affect the melt flow index (MFI) of the bio-composite compound. The tendency of the melt flow index is the same trend as the previous part. The addition of the LE powder into the matrix causing the MFI was increasing shown in Figure 49.

The neat PLA compound has MFI as 5.83 g/10min. When adding the PBS affects the bio-polymer blend compound can be easier to melt flow which increasing the MFI value to 13.85 g/10min. Which the reason ever pronounces in the previous part that the structure of PBS has a long chain more than the PLA chain. Especially, PBS is the flexible polymer. (7) Thus, PBS bind with the PLA matrix affects the chain mobility of the polymer compound thus easier to flow and increasing MFI value. The addition of the LE powder into the matrix affect the MFI was increasing more than the neat PLA compound and L90\_B10 compound. The lowest content of LE powder has the MFI increasing to 23.87 g/10min and the MFI value increasing to 47.52 g/10

min with the content of LE powder as 10%w/w. The increasing the MFI trend with the greater content of LE powder because the LE particles were bidden in the matrix causing reduction the continuous phase of the matrix. Moreover, LE may be reduce the entanglements of polymer chain affects MFI of the bio-composite compound has increased. The results were related to the SEM images of the cross-section morphology of the bio-composite compound that shown in Table 20. The interaction between the matrix and LE particles has a low interaction because some particles of LE powder were pulled out of the matrix.



**Figure 49 Melt flow index (MFI) of the neat PLA compound and the bio-composite compound**

## **2. Bio-composite film from PLA, PBS, and LE powder**

The bio-composite film based on PLA incorporated PBS and LE powder were performed by the blow film extruder. The bio-composite film has a thickness of 70-80 micron and a width of 16-17 cm. The influence of the addition of PBS and the LE powder as fillers into the PLA matrix were discussed. Characterization of the bio-composite films was examined the chemical properties (FT-IR and XRD), mechanical properties (Tensile testing), physical properties (SEM, Color parameter, UV-transmittance, and Water vapor permeability), and field test by cover the soil during in the environment.



### **The chemical properties of the bio-composite film**

The neat PLA film, the bio-polymer blend film, and the bio-composite films were to analyze the chemical properties by Fourier Transform Infrared Spectroscopy (FT-IR) and X-ray differential (XRD). The effect of the LE powder that causing the bio-composite film was how changed in the chemical properties were studied.

#### **1. Fourier Transform Infrared Spectroscopy (FT-IR)**

The film specimen and LE powder were to determine the functional group by using Fourier Transform Infrared Spectroscopy (FT-IR) with ATR-mode. Which scan in the wavenumber ranges on 400-4000  $\text{cm}^{-1}$ . Functional group of the bio-composite film when adding with the PBS and LE powder were observed. Distinct of the functional group of the neat PLA film, the bio-polymer blend film, and the bio-composite film were interpreted.

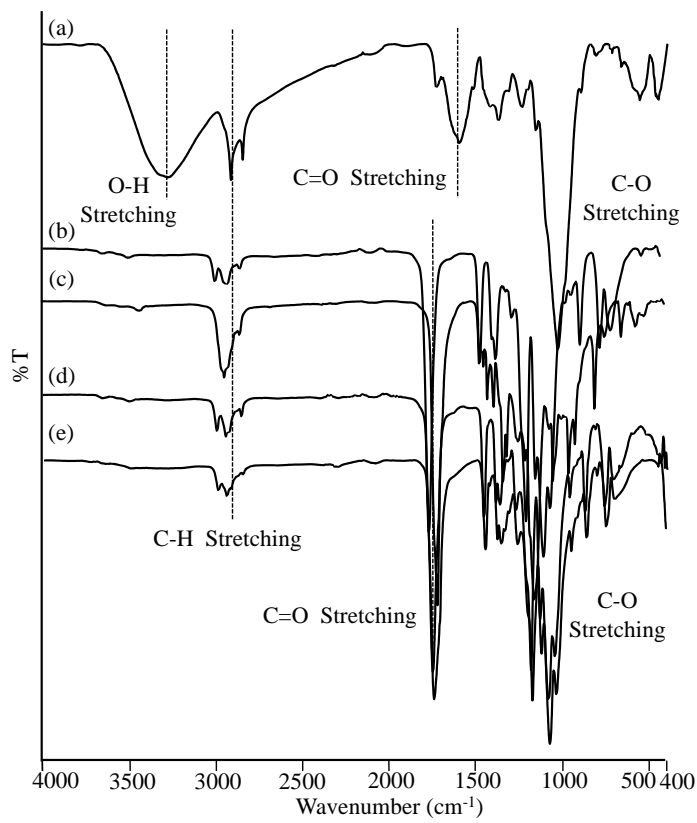
The FT-IR data of the LE powder, the neat PLA film, the neat BS film, the bio-polymer blend film, and the bio-composite film were promoted in Table 21. The LE powder has the 4 main peaks at 3281.69  $\text{cm}^{-1}$ , 2961.88  $\text{cm}^{-1}$ , 1599.92  $\text{cm}^{-1}$ , and 1031.49  $\text{cm}^{-1}$ . The interpretation was determined O-H stretching, C-H stretching, C=O stretching, and C-O stretching, respectively. The functional groups of LE powder related to the composition of lemongrass leaf as cellulose, hemicellulose, and lignin. (47, 56, 57) The neat PLA film has 3 main peaks as C-H stretching at 2994.01  $\text{cm}^{-1}$ , C=O stretching at 1745.66  $\text{cm}^{-1}$ , and C-O stretching at 1077.61  $\text{cm}^{-1}$ . All spectra appear of the neat PLA film were shown the functional groups follow the structure of poly(lactic acid). (7) The neat PBS film has 3 main peaks like PLA spectrum. The interpretation peak can separate at 2945.35  $\text{cm}^{-1}$  as C-H stretching, at 1711.83  $\text{cm}^{-1}$  as C=O stretching, and, at 1152.14  $\text{cm}^{-1}$  as C-O stretching. The addition of PBS content into the PLA matrix also presents 3 main peaks but each spectrum was slightly shifted to a higher wavenumber. The shift of spectrum can observe in Figure 50. The functional groups of the bio-polymer blend films also according to the chemical structure of poly(lactic acid) and polybutylene succinate. (7)

The additional content of LE powder into the bio-polymer blend film also available has 3 main peaks include C-H stretching, C=O stretching, and C-O stretching. The spectrum of the bio-composite film was found at the same peak with

the neat PLA film and the bio-polymer blend film that just a slight shift of wavenumber. The FT-IR spectrum of the bio-composite film non-appear the functional group with O-H stretching, which is the main peak of LE powder. When compared the ratio of the matrix and LE powder, it found that the content of LE powder has less than the matrix. Detection of instrument was easily to determine the main material more than the LE particles. Thus, the FT-IR spectrum of the bio-composite was an invisible peak of O-H stretching. Moreover, this work was analyzed by both FT-IR and ATR modes. This mode was analyzed only the surface of the specimen. (66) The surface specimen of the bio-composite film has not found the particle of LE which the bio-composite film has a roughness but not found the LE particles pulled out to surface specimen. It can observe the characteristic with SEM image of the bio-composite film in Table 22. So, the attribute is another reason for explaining about the FT-IR spectra of the bio-composite film non-found the O-H stretching peak.

**Table 21 FT-IR data of natural fillers and the bio-composite film**

Samples	Wavenumber (cm <sup>-1</sup> )			
	O-H Stretching	C-H Stretching	C=O Stretching	C-O Stretching
Natural fillers				
LE powder	3281.69	2961.88	1599.92	1031.49
Films				
Neat PLA film	-	2994.01	1745.66	1077.61
Neat PBS film	-	2945.35	1711.83	1152.14
Bio-polymer blend film	-	2994.44	1746.88	1078.76
Bio-composite film	-	2993.99	1747.90	1079.44



**Figure 50** FT-IR spectrum of (a) lemongrass leaf powder, (b) neat PLA film, (c) neat PBS film, (d) bio-polymer blend film, and (e) the bio-composite film (B10\_LE)

## 2. X-ray diffraction (XRD)

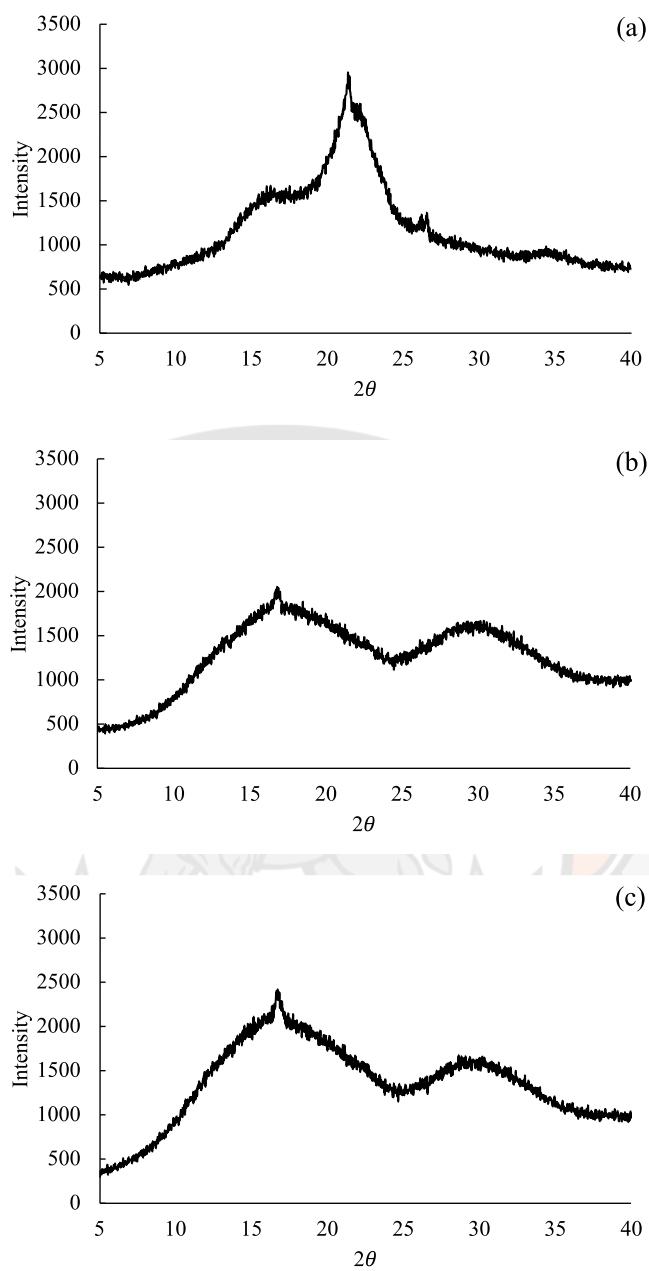
The bio-composite film specimen and LE powder were used to determine the crystallinity and amorphous by using X-ray diffraction (XRD). The effect of the LE powder on the crystallinity or amorphous change in the bio-composite film were studied.

The diffractogram of LE powder was presented in Figure 51. The LE powder has a main peak of  $2\theta$  at  $16.7$  and  $21.48^\circ$ , which relates to the cellulose of lemongrass leaf. However, the peak of LE powder presents a rather broad peak thus express the amorphous in lemongrass leaf. In the study of Olopade and co-worker was reported the lemongrass is the perennial grass that has silica content. The leaf of perennial grass has a high content of silica in the leaf. (67-69) So, this work was used

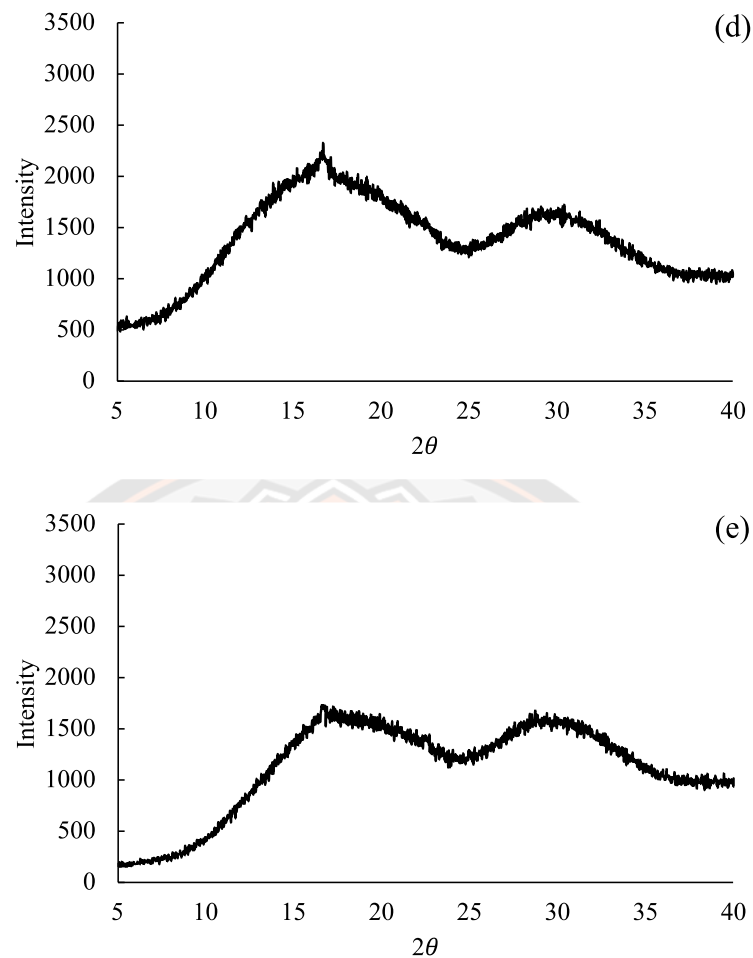
the lemongrass leaf thus the XRD pattern was expressed the amorphous peak of amorphous silicon. The XRD pattern of the neat PLA film and the bio-polymer blend film were appeared the broad band of the amorphous. The neat PLA film and the bio-polymer blend film have 2 main peaks include around  $2\theta = 16^\circ$  and  $30^\circ$ . Adding the PBS content into the matrix has decreased slightly intensity of the amorphous peak. Furthermore, addition of LE powder into the bio-polymer blend matrix caused the intensity of the peak of bio-composite film was slightly changed.

The diffractograms of all bio-composite films appeared the broad band same the neat PLA film and the bio-polymer blend film. The XRD pattern of all bio-composite films was found 2 mains peaks of  $2\theta$  consists of approximately  $16^\circ$  and  $30^\circ$  which non-different from the neat PLA film and the bio-polymer blend film. The bio-composite film has not expressed the peak of LE powder in the XRD pattern. The diffractogram of the bio-composite film was shown in Figure 51. The increasing of LE powder into the bio-polymer blend matrix affects the intensity of the  $2\theta$  peak was changed, when increase LE powder to high content causing the intensity was decreased.

In the discussion, when increase LE powder that affects decreasing of amorphous phase in bio-composite film, which can confirm by  $2\theta$  peak intensity in Figure 51d-e was decreased. The lemongrass leaf also has the content of crystalline around 36.20%. (69) So, the addition over the content of LE powder maybe affects decreasing the amorphous phase in the bio-composite film.



**Figure 51 XRD pattern of (a) LE powder (b) B10\_LE2.5, (c) B10\_LE5.0, (d) B10\_LE7.5, and (e) B10\_LE10.0**



**Figure 51 (Cont.)**

### **The morphology of the bio-composite film**

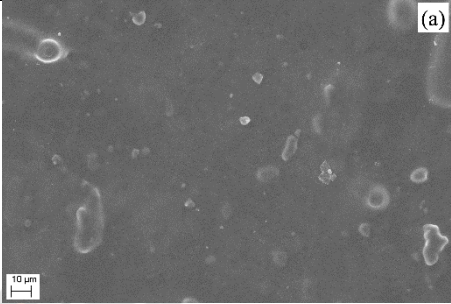
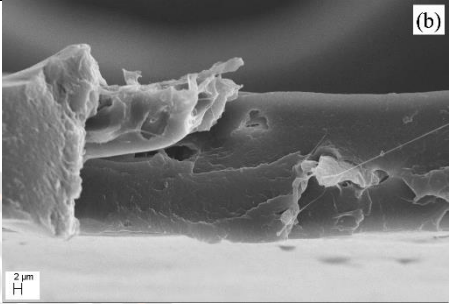
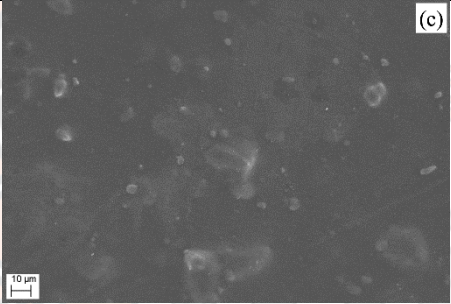
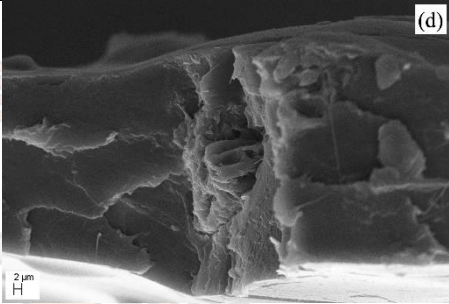
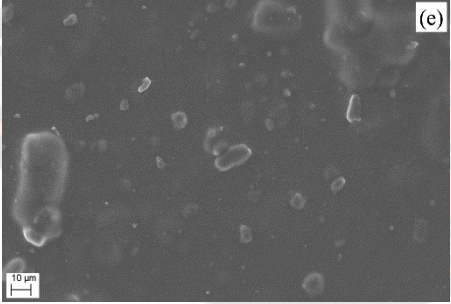
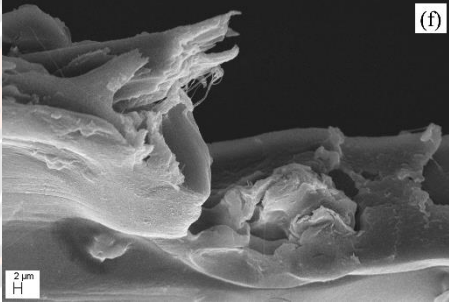
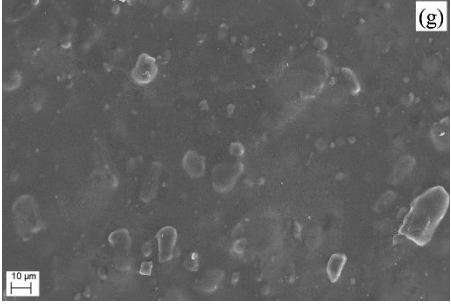
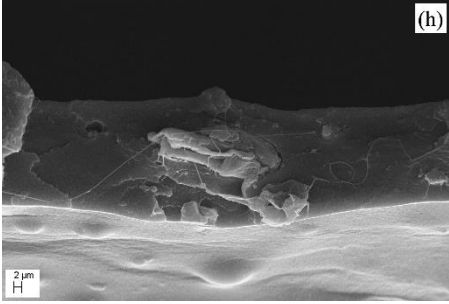
All specimens of the neat PLA film, the bio-polymer blend film, and the bio-composite film were to characterize the morphology using scanning electron microscopy (SEM). The evaluation at the magnification of 200X was used to observe the surface morphology. For 1000X as magnification to study the cross-section morphology and interaction between matrix and LE particles. The specimen of the cross-section for determination in SEM was received after tensile testing. The effect of tension force on the interaction during matrix and fillers was discussed.

Table 22 was assigned the surface morphology and the cross-section morphology of the bio-composite film after tensile testing. The surface morphology of the bio-composite film has a roughness that increasing roughness when adding more content of the LE powder. The LE particles are also bidden in the bio-composite film

which non-pulled out to the surface. The LE powder has a particle with different shapes that LE particles express the rod shape and flake. The different shapes and sizes have the advantage in the distribution in the bio-polymer blend matrix. A small particle can disperse and fill between any particles in the bio-polymer blend matrix. If the LE particles have only size and shape causing the LE particles were generated more agglomerate. The probably poor distribution in the bio-polymer blend matrix affects the properties of the bio-composite film. However, the oversize of LE particles influences the bio-composite film has roughness because the LE particle was distributed and convex to the film surface.

The cross-section of the neat PLA film, the bio-polymer blend film, and the bio-composite film from tensile testing was observed in Figure 43-45 in previous part. The cross-section surface morphology of the neat PLA film shows a continuous phase that the specimen was slightly stretch after received the tension force. The addition of the PBS into the PLA matrix expresses the cross-section morphology after carried on the tension force has a smooth surface less than the neat PLA film. However, the cross-section morphology also has a continuous phase and the phase non-separated between PLA and PBS. While the addition of the LE powder into the bio-polymer blend matrix was shown the LE particle was bidden in the matrix phase. The tension force has affected the bio-composite film that the LE particle was pulled by the tension force and pulled out from the matrix. But some particles of LE also embedded in the matrix, which can see in Table 22d. Moreover, it was found the gap between the LE particle and the matrix is less gap and non-found the hole on the cross-section morphology. It can be discussed that the matrix and LE particle has a good interaction because the LE particle has the rough surface thus causing the matrix can involve with the rough surface. However, increasing of LE particle for adding into the matrix that causing to reduce the continuous phase that maybe reduces the mechanical properties of the bio-composite film.

**Table 22 Surface Morphology and Cross-section of the bio-composite film after tensile testing**

Sample	Magnification	
	Surface at 200X	Cross-section at 1000X
B10_LE2.5		
B10_LE5.0		
B10_LE7.5		
B10_LE10.0		



### **The mechanical properties of the bio-composite film**

The mechanical properties of the bio-composite films were examined by tensile testing. Bio-composite film in 2 directions consists of mechanical direction (MD) and transverse direction (TD) were tested by Tensile testing. The effect of LE powder adding into the bio-polymer blend matrix on the mechanical properties of the bio-composite films were characterized and studied. The results were reported as tensile strength at break, %elongation at break, and modulus at break.

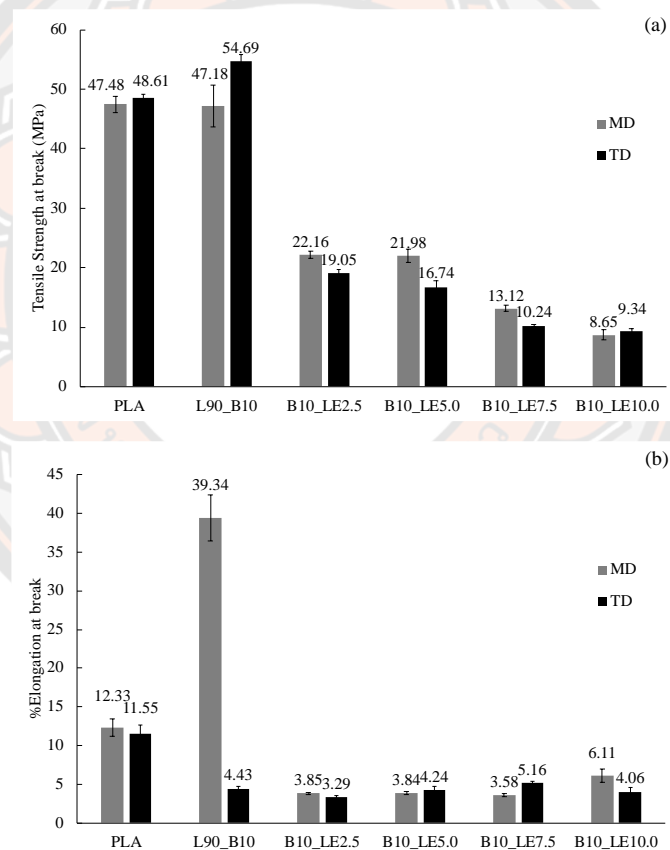
Figure 52 was exhibited the mechanical properties of the neat PLA film, the bio-polymer blend film, and the bio-composite film with various content of LE powder. The neat PLA film has the tensile strength at break as 47.48 MPa in MD and 48.61 MPa in TD. The addition of PBS at 10%w/w content causing the bio-polymer blend film has increased the value of tensile strength. In the above part on the mechanical properties of the bio-polymer blend film were discussed. The addition of LE powder into the bio-polymer blend matrix was changed the tensile strength decrease. The tendency of the LE powder into the bio-polymer blend matrix was presented the decreasing trend when the greater content of LE powder. The addition of LE powder at the lowest content as 2.5%w/w was found the tensile strength has decreased to 19 MPa in MD and 22 MPa in TD. The increasing LE powder to the highest content as 7.5%w/w causing reduce the tensile strength at break to 8 MPa in MD and 9 MPa in TD. The reduction trend in tensile strength at break when increasing content of LE powder due to LE particles were distributed and embedded in the bio-polymer blend matrix. Because LE particles reduced the continuous phase of the matrix when the specimen received the tension force affect the surface between LE particle and matrix can be easier to separate. So, adding over the content of LE powder into the matrix causing the bio-composite film has more LE particles in the matrix. Moreover, the observation of the SEM image was found the area between LE particles and the matrix have a little gap and voids. Because LE particles have increased the opportunity was to pull out and separated, which can be observed by the cross-section morphology of the bio-composite film after tensile testing and the interaction between LE particles and the matrix in Figure 52.

%Elongation at break of the neat PLA film, the bio-polymer blend film, and the bio-composite film has appeared in Figure 52. %Elongation at break of the

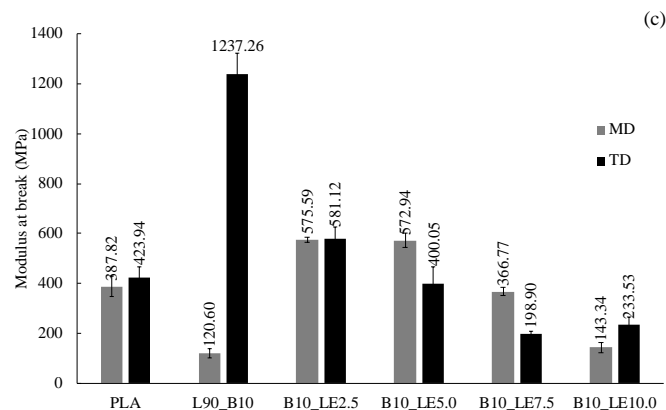
neat PLA film has around 11% in MD and 12% in TD. The adding of PBS content causing %elongation at break has the broad ranges of around 4% in MD and 39% in TD that has a slightly more than the neat PLA film. The discussion of this case may be the effect of the PBS content can improve the strain of the PLA. Which can follow the discussion on the above part in the mechanical properties of the bio-polymer blend film. While adding content of LE powder into the matrix causing the %elongation at break was decreased. The trend of %elongation of the bio-composite film when varies content of LE powder from 2.5-10.0%w/w was shown a slightly increasing of %elongation. The lowest content of LE powder in the bio-composite film has %elongation at break was decreased to 3% in MD and 4% in TD. The increasing content of LE powder has exhibited %elongation at break was improved to 4% in MD 6% in TD at the highest content of LE powder. This behavior has occurred because of the effect of PBS content adding in the matrix. It can improve the strain of the bio-composite film. The more content of LE particle maybe can be involved with the polymer chain thus effects the polymer chain in the matrix can be rearranged. The specimen of the bio-composite film carried on the tension force causing LE particle was involved and pulled the polymer chain in the polymer matrix and increasing of strain. However, the addition of LE powder influences was reduced the continuous phase of the bio-polymer blend matrix that has %elongation was less than the neat PLA film and the bio-polymer blend film. Modulus at break of the neat PLA film, the bio-polymer blend film, and the bio-composite film can be observed in Figure 52. The modulus at break of all bio-composite film has decreased with the greater content of LE powder adding. In the discussion, general the PLA was hard, and brittle properties thus has a high modulus. (7) The adding of PBS can slightly improve the flexibility of the PLA. However, the addition of LE powder into the matrix cannot improve the flexibility of PLA which has a high modulus at break.

In the summary, the addition of the LE powder into the bio-polymer blend matrix was affected to reduce the continuous phase of the bio-polymer blend matrix. Thus, it can reduce the strength of the bio-composite film. This result can be observed by the SEM image. In Table 22 was shown the LE particle was hidden in the matrix. The bio-composite film with the LE powder has immiscibility because the matrix and LE particle were adhesive with poor interaction which shows the gap between LE

particle and matrix. Moreover, the tensile strength in the machine direction is more than the transverse direction because the film was extended in the machine direction cause the polymer chain rearrangement that affects to high strength in MD more than TD. The mechanical results can be concluded the addition of LE powder causing the bio-composite film comes to an incompatibility. Because the LE powder was reduced the tensile strength and decrease strength high to 40% when compared with the bio-polymer blend film. It can be explained the LE powder is non-reinforcement. However, the addition of the LE powder has the advantage of the color parameter and UV-light absorption.



**Figure 52 Mechanical properties of the bio-composite film as (a) tensile strength at break, (b) %elongation at break, and (c) Modulus at break**



**Figure 52 (Cont.)**

### The physical properties of the bio-composite film

The physical properties of the bio-composite film were characterized by water vapor permeability (WVP), color parameter, and UV transmittance. PBS content and various content of LE powder how affects the physical properties of the bio-composite film were studied.

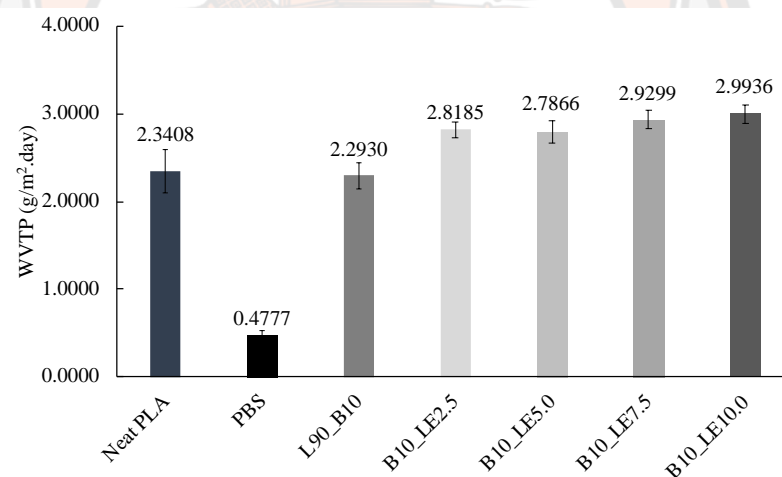
#### 1. Water vapor permeability (WVP)

The water vapor permeability was using test the water vapor transmission (WVTR) of the bio-composite film which follows ASTM E96-80. The content of PBS and various content of LE powder for adding which affect water vapor transmission through the bio-composite film were determined. The bio-composite films were applied to produce the mulch film to cover the soil. Thus, this is an important property of the mulch film for keep and control the moisture in the soil.

Figure 53 was notifying the water vapor transmission (WVTR) of the neat PLA film, the bio-polymer blend film, and the bio-composite film. The WVTR of the neat PLA film has 2.3408 g/m<sup>2</sup>·day, while in the neat PBS film has 0.4777 g/m<sup>2</sup>·day. The adding content of PBS has a slight decrease of WVTR because the addition of PBS helps the polymer chain of the bio-polymer blend has good orientation and good entanglement. The addition of LE powder into the bio-polymer blend matrix causing the WVTR to have an increasing trend with the greater content of LE powder. WVTR of the bio-composite film has a range of 2.8000-3.0000

$\text{g/m}^2\cdot\text{day}$  which has WVTR nearly with the neat PLA film and the bio-polymer blend film.

The observation of the cross-section morphology with SEM image were shown in Table 22. The addition of LE powder affects the surface has a roughness. Some particles of LE were convex on the film surface that may generate a small pore on the surface. The high content of LE powder has reduced the continuous phase and generated the agglomerate in the matrix. Moreover, the agglomerates of natural fillers were poorly distributed affect to the interaction between natural particles and PLA and PBS chain that the cross-section surface of the bio-composite film shows a gap and voids. Because the vapor can be diffused through out the bio-composite film. (37, 59) So, the WVTR tendency thus increasing when adding over the content of LE powder in the bio-polymer blend matrix.



**Figure 53 Water vapor transmission (WVTR) of the neat PLA film, the neat PBS film, the bio-polymer blend film, and the bio-composite film with various contents of LE powder**

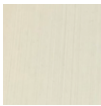


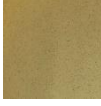

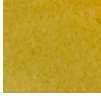
## 2. Color parameter

The color parameter was explained the different color parameters of the bio-composite film compared to the neat PLA film and the bio-polymer blend film. The result was reported the color parameter consists of L value meaning the lightness-darkness of the specimen,  $a^*$  value is red/green shade, and last is  $b^*$  value assigned to yellow/blue shade. Moreover, the  $\Delta E$  was indicated about the changing the color parameter of the bio-composite film compared to the neat PLA film and the bio-polymer blend film.

The color parameters of the neat PLA film, the bio-polymer blend film, and the bio-composite film were indicated in Table 23. The neat PLA film and the bio-polymer blend film have translucent which has a high value of the color parameter. Which the bio-polymer blend film has almost no different from the neat PLA film. The data of color parameters show  $\Delta E$  of the bio-polymer blend film has just 1.20 when compared with the neat PLA film.

The bio-composite film incorporated with LE powder causing the color parameters were changed which the addition of LE powder has reduced the lightness of the film. L value was reducing and increasing values of  $a^*$  and  $b^*$ . The increasing content of LE powder causing the  $\Delta E$  of bio-composite film was changed to a high value. The bio-composite film with LE powder appeared in a green-yellow shade that following the shade of lemongrass leaf. The lemongrass leaf has a green-yellow shade because the main pigment in the lemongrass leaf is chlorophyll A and chlorophyll B. Which, blue-green pigments are chlorophyll A and chlorophyll B has yellow-green pigments. (49) The addition of LE powder can improve the colorlessness of the bio-polymer blend film, which the color of the bio-composite film has the advantage of resistance the UV-light transmittance. Because avoid weed growth because the grass must use UV-light for photosynthesis. (55) Component of the pigment as chlorophyll in the LE powder can absorb UV-light. (61) Thus, affect the grass and weed were reduced growth.

**Table 23** The color parameter of the neat PLA film, the bio-polymer blend film, and the bio-composite film

Samples	Color parameters			$\Delta E$	
	L	a*	b*		
Neat PLA film	$86.89 \pm 0.34$	$-0.20 \pm 0.00$	$-4.67 \pm 0.07$	Ref.	
L90_B10	$87.11 \pm 0.07$	$-1.20 \pm 0.00$	$-4.05 \pm 0.05$	1.20	
B10_LE2.5	$83.97 \pm 0.33$	$0.45 \pm 0.08$	$4.59 \pm 0.47$	9.73	
B10_LE5.0	$81.00 \pm 0.63$	$0.80 \pm 0.12$	$11.38 \pm 0.70$	17.13	
B10_LE7.5	$78.35 \pm 0.60$	$1.41 \pm 0.16$	$13.88 \pm 0.81$	20.48	
B10_LE10.0	$76.90 \pm 0.87$	$2.19 \pm 0.30$	$15.11 \pm 1.03$	22.29	

### 3. UV Transmittance

UV transmittance was exported data as %UV transmittance (%T) in the visible region. The result was reported in 3 regions that identify at 315 nm (UV-B), 400 nm (UV-A), and 700 nm (visible region). Absorption ability of the LE powder when was adding into the bio-composite film was studied.

%UV transmittance data of the neat PLA film, the bio-polymer blend film, and the bio-composite film were analyzed in Table 18. The neat PLA film has the highest %T as high as 81-90%. Due to the neat PLA film has clear and colorless. So, the UV-light can be easier to transmit through the neat PLA film. The bio-polymer blend film has %T less than the neat PLA film. Generally, PBS is a non-transparency polymer. (7) Thus, adding the content of PBS into the PLA matrix

causing the bio-polymer blend film appears rather non-transparent. Which the UV-light can transmit through the bio-polymer blend film was reduced to 70-88% from 81-90% of the neat PLA film. Both the neat PLA film and the bio-polymer blend film cannot protect UV-light.

%T of the bio-composite film was decreased when incorporated with the greater content of LE powder. The data of the bio-composite film with various content of LE powder has performed in Table 24. The lowest content of LE powder adding into the bio-polymer blend film has %T was decreased to 13-23%. While the adding with the highest content of LE powder that influence reduce %T of the bio-composite film to 1-3%. Which the bio-composite film with 10%w/w of LE powder can absorb the UV-light higher than 95%. The bio-composite film with the LE powder has UV protection ability. The addition of LE powder into the bio-composite film can improve the UV protection due to the LE particles were distributed in the matrix act as the UV protection barrier. Because the LE powder has the UV absorption compounds as chlorophyll A and chlorophyll B. Which the chlorophyll A can highly absorb UV-light ability in regions 450 and 680 nm. The chlorophyll B has the highest absorb UV-light ability in wavelength 480 nm. (55, 61) Moreover, the LE powder also has the aromatic compound that can greatly absorb the UV-light consist of phenols, flavonoids, and alkaloids. (52, 62, 63) So, the addition of LE powder was improved to reduce the UV-light transmit through the bio-composite film to the soil.

In the summary, the bio-composite film incorporated with LE powder can absorb UV-light more than 95%. The LE powder act as the barrier to prevent the UV-light, which the bio-composite film with less content of LE powder has translucent. The increasing of LE powder causing the bio-composite film to come to the non-translucent film. The bio-composite film in this work was applied to produce the mulch film. So, using the bio-composite film to cover the soil causing reduce the grass and weed growth in the plot. The grass and weed growth depend on UV-light for photosynthesis which photosynthesis by UV-visible light. (60) When using the bio-composite film to cover the soil that affects reduces the UV-light transmit through to the grass and weed in the soil because of prevention the growth of grass and weed in the plot. This is an advantage to avoid herbicides to remove grass and weed and reduce the toxic chemical release to the environment.



**Table 24 %UV-transmittance of the neat PLA film, the bio-polymer blend film, and the bio-composite film**

Samples	%UV-transmittance		
	at 315 nm (UV-B)	at 400 nm (UV-A)	at 700 nm (Visible region)
Neat PLA film	81.00 ± 1.00	85.33 ± 0.58	90.00 ± 0.00
L90_B10	70.00 ± 2.65	79.33 ± 2.08	88.33 ± 1.15
B10_LE2.5	13.67 ± 1.15	17.33 ± 0.58	23.33 ± 0.58
B10_LE5.0	4.00 ± 0.00	4.67 ± 0.58	8.00 ± 0.00
B10_LE7.5	2.33 ± 0.58	3.33 ± 0.58	6.00 ± 0.00
B10_LE10.0	1.00 ± 0.00	1.00 ± 0.00	3.00 ± 0.00

#### **Thermal properties of bio-composite film**

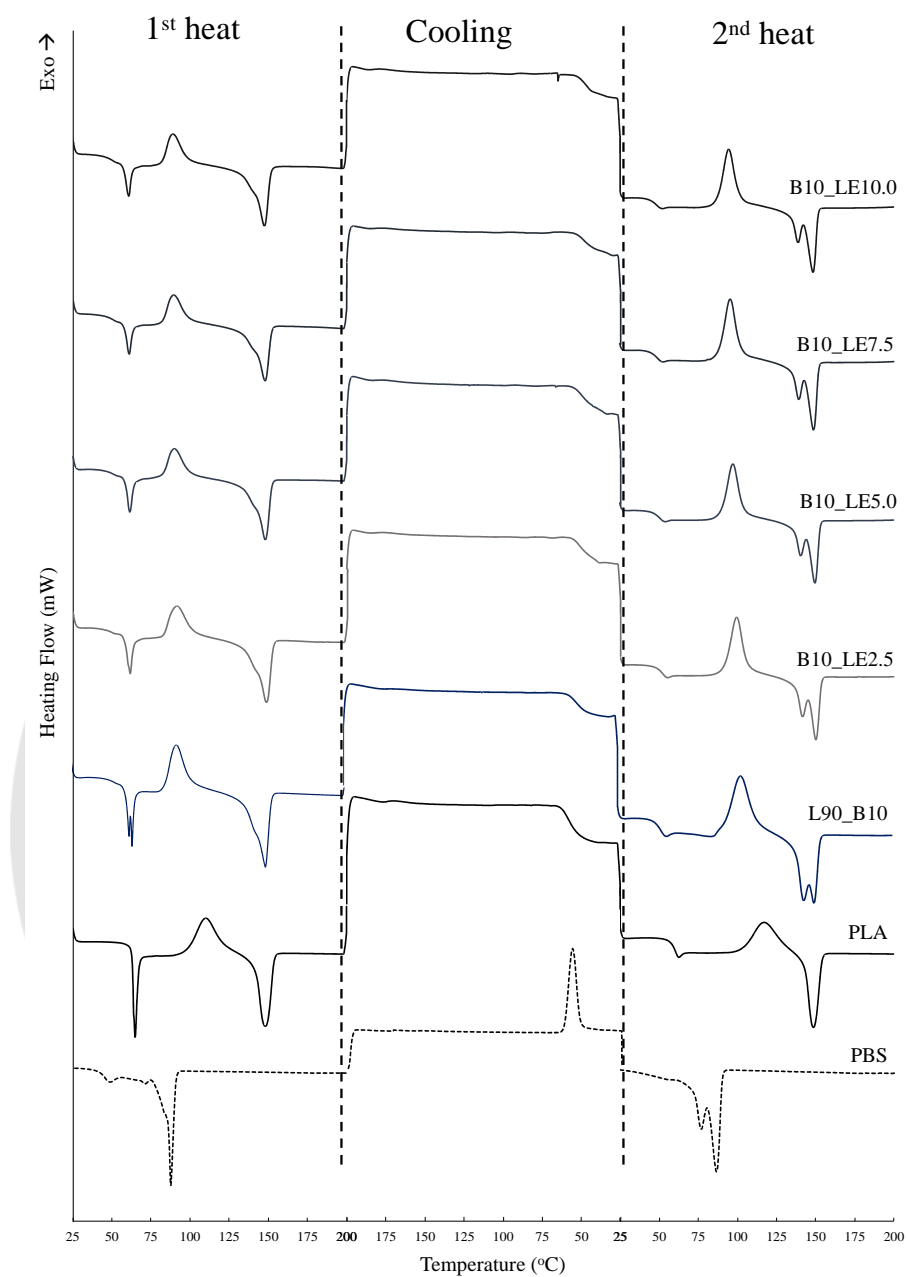
Differential scanning calorimeter (DSC) was used to determine the thermal properties of the neat PLA film, the bio-polymer blend film, and the bio-composite film. The data from the DSC thermogram was reported with the second heating consists of glass temperature ( $T_g$ ), the cold crystallization temperature ( $T_{cc}$ ), crystalline melting temperature ( $T_m$ ), enthalpy of melting ( $\Delta H_m$ ), and the degree of crystallinity ( $\%X_c$ ). The miscibility of the bio-composite film when adding PBS content and LE powder was studied.

Table 25 shows the data from the DSC thermogram of the neat PLA film, the bio-polymer blend film, and the bio-composite film.  $T_g$  of the neat PLA film was presented 57.04 °C, while the neat PBS film was not found  $T_g$ . The addition of PBS content causing  $T_g$  of the bio-polymer blend film has changed to a lower temperature which appears at 48.07 °C.  $T_g$  of the bio-composite film incorporated with various content of LE powder were shifted to a lower temperature with the greater content of LE powder. At the lowest content of LE powder was found that  $T_g$  peak of the bio-polymer blend film shows was slightly shift from 48.07 °C to 47.28 °C. The increasing to highest content of LE powder affects to  $T_g$  was shifted to a lower temperature to 43.36 °C that shown in the DSC thermogram in Figure 54.

$T_{cc}$  of the neat PLA was detected at 103.18 °C, while in PBS is not found the  $T_{cc}$  peak. The bio-polymer blend film shows  $T_{cc}$  peak that was shifted to a lower temperature and appears around 92.34 °C. The addition of LE powder into the bio-composite film affects  $T_{cc}$  was appeared faster than the neat PLA film and bio-polymer blend film.  $T_{cc}$  onset of the bio-composite film was started at 91.50 °C with 2.5%w/w of LE powder. The increasing content of LE powder to 10%w/w influences  $T_{cc}$  has a shift to lower temperature more than the less content of LE powder.  $T_{cc}$  of B10\_LE10.0 is detected at 87.23 °C. The effect of addition of PBS content and LE powder on  $T_{cc}$  were shifted to the lower temperature that acts as nucleating agent.(65, 70)

$T_m$  of the neat PLA film was promoted around 142.82 °C.  $T_m$  peak of the neat PLA film.  $T_m$  of PBS shows 72.84 °C. The addition of PBS causing  $T_m$  peak of the bio-polymer blend film has a double peak. It was found  $T_m$  peak has approximately 141.78 °C. All of bio-composite film has  $T_m$  ranges on 141-144 °C. The addition of LE powder affects  $T_m$  of the bio-composite film has two distinct peaks. The melting temperature of the bio-composite film has two distinct peaks that cause from the immiscibility of LE powder and polymer chain, which relates to the SEM image of the bio-composite compound and the bio-composite film show the immiscibility morphology.  $\Delta H_m$  of the bio-composite film has value more than the neat PLA film and the bio-polymer blend film. Which influences %crystallinity (% $X_c$ ) of the bio-composite film.

%Crystallinity (% $X_c$ ) of the neat PLA film, neat PBS film, bio-polymer blend film, and bio-composite film were shown in Table 25. The neat PLA film has 22.95% and the bio-polymer blend film has % $X_c$  more than the neat PLA film. The addition of LE powder causing % $X_c$  was increased but % $X_c$  of bio-composite films are not different when increasing LE powder. The addition of LE powder affects to a slightly increasing of % $X_c$  that can also discuss the LE powder is probably the nucleating agent. (70, 71) The results relate to XRD of the bio-composite film as the increasing of LE powder affects the bio-composite film promoted the decreased intensity of the amorphous peak.



**Figure 54 DSC thermograms of the neat PLA film, the bio-polymer blend film, and the bio-composite film in second heating**

**Table 25 Data of DSC analysis of the neat PLA film, the bio-polymer blend film, and the bio-composite film**

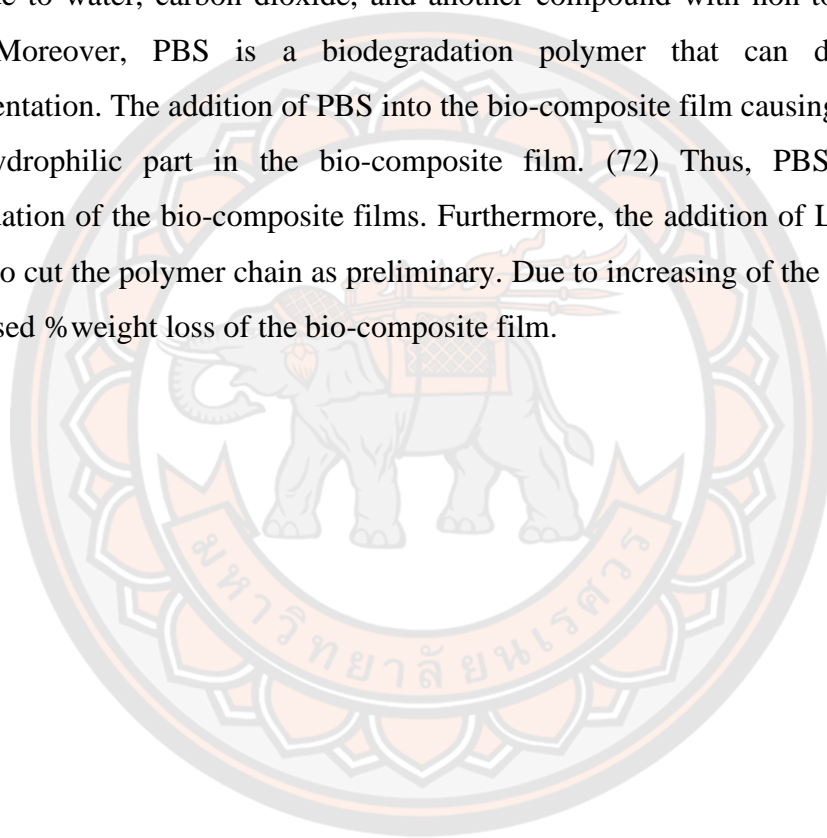
Sample	2 <sup>nd</sup> heat				
	T <sub>g</sub>	T <sub>cc</sub>	T <sub>m</sub>	ΔH <sub>m</sub>	%X <sub>c</sub>
Neat PLA	57.04	103.18	142.82	21.34	22.95
PBS	N/A	N/A	72.84	21.11	10.05
L90_B10	48.07	92.34	141.78	27.09	32.37
B10_LE2.5	47.28	91.50	144.28	29.72	36.52
B10_LE5.0	46.01	89.68	143.58	30.87	39.05
B10_LE7.5	44.22	88.13	142.40	30.83	40.18
B10_LE10.0	43.46	87.23	141.77	31.36	42.15

#### **The biodegradability of the bio-composite film**

The neat PLA film, the bio-polymer blend film, and the bio-composite film were to assay the biodegradability via embed the specimen in the soil for 2 months, which the report was present %weight loss every month. The effect of PBS content and LE powder in the bio-composite film on the decomposition was investigated.

The film samples before putting in the soil and the specimen recovering after embedding in the soil for 1 and 2 months were exported in Table 26. The whole specimen recovering also were remains the appearance, non-cracking. The trend of %weight loss was represented in Figure 55. Which the measurement of weight loss of the whole specimen was found the neat PLA film has the lowest of weight loss. %Weight loss of the neat PLA film was a slight loss of just 0.21% in 2 months. While in the neat PBS has high %weight loss around of 31.25%. The addition of PBS content affects %weight loss of the bio-polymer blend film loss to 0.46% in 2 months. The addition of LE powder into the bio-composite film affects the bio-composite film has more degradation. %Weight loss of the bio-composite film promising increase with the greater content of LE powder. The highest %weight loss is the bio-composite film with 10%w/w of LE powder which can degrade to 2.50% for 2 months.

The observation in the SEM image promoted the bio-composite film with the LE powder that has miscibility. LE particles were distributed, imbed, and reduce the continuous phase because of reducing the strength of the specimen. So, the specimen were tested the biodegradability by burying in the soil causing moisture and microorganism can be easily to cut the polymer chain. Because PLA can decompose by hydrolysis process which includes the moisture, microorganism, and content of oxygen was influenced the degradation of PLA. Then decomposition, PLA will degrade to water, carbon dioxide, and another compound with non-toxic chemicals. (33) Moreover, PBS is a biodegradation polymer that can degradation by fragmentation. The addition of PBS into the bio-composite film causing an increase in the hydrophilic part in the bio-composite film. (72) Thus, PBS can improve degradation of the bio-composite films. Furthermore, the addition of LE powder thus helps to cut the polymer chain as preliminary. Due to increasing of the LE powder has increased %weight loss of the bio-composite film.



**Table 26 The bio-composite film sample recovered after biodegradable in soil**

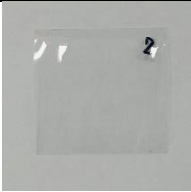
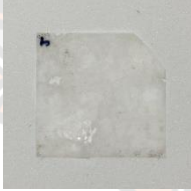
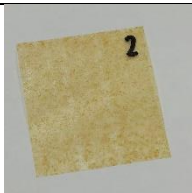

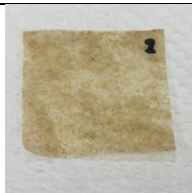
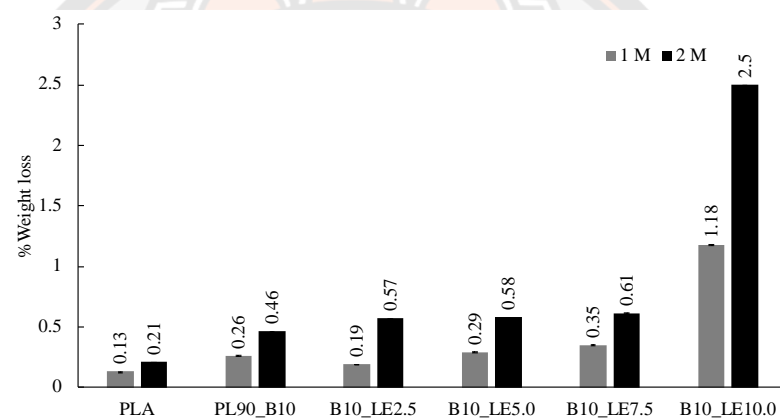
Sample	Biodegradation times (months)		
	Start	1	2
Neat PLA film			
Neat PBS film			
L90_B10			
B10_LE2.5			
B10_LE5.0			
B10_LE7.5			

Table 26 (Cont.)

Sample	Biodegradation times (months)		
	Start	1	2
B10_LE10.0			



**Figure 55 Biodegradability of the neat PLA film, the bio-polymer blend film, and the bio-composite film**

#### Field test of the bio-composite film

Field testing is a model with the real conditions for using the mulch film. Lead the neat PLA film, the bio-polymer blend film, and the bio-composite film were to cover the soil during the environment. The whole specimen was to cover on the soil for 2 months which follow with the cultivation time. Physical properties (color parameter and UV transmittance) and the mechanical properties by tensile testing were discussed.



















Figure 56 presents the color parameter ( $L^*$ ,  $a^*$ , and  $b^*$ ) of the neat PLA film, the bio-polymer blend film, and the bio-composite film after field test by the cover on the soil. The result shows all the film were changed to be lightness by

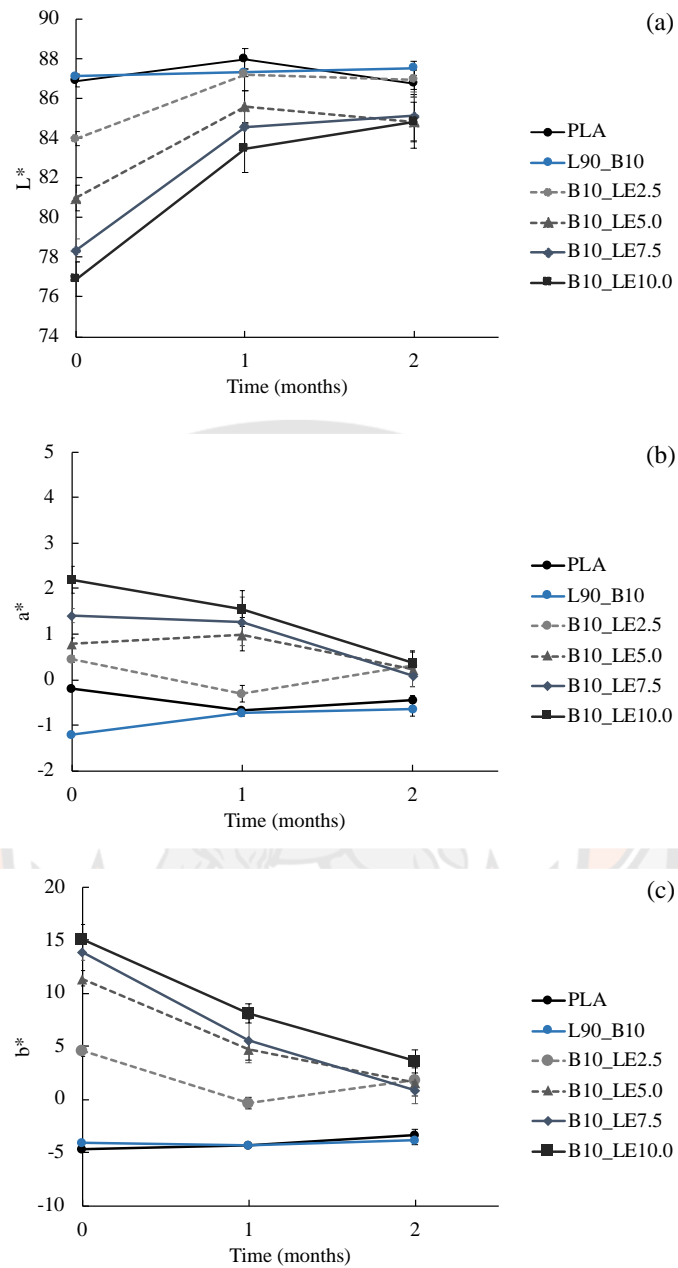
increasing in  $L^*$  value. The values of  $a^*$  and  $b^*$  were decreased to a low value when time passed. Because  $\Delta E$  was changed to low value, which the color of film was become pale. Thus, It has the color nearly with the neat PLA film, which can see the color change of the film specimen after field test in Table 27.

The color parameter and tone of the neat PLA film and the bio-polymer blend film have remained the value and shade non-different from the specimen before the field test. However, bio-composite film was increasing in lightness and become pale when covering the soil for a long time. The bio-composite film has changed the tone of color may be from the LE powder. The chlorophyll is the pigment in the lemongrass leaf. This compound can react with oxygen and moisture in the air. When the color compound reacts with oxygen and moisture then maybe changes the structure and causing the color of the compound was changed which, affects the bio-composite film thus has a different color shade. (73) Moreover, the chlorophyll can be degraded by the light which the compound can absorb the light and over absorption become unstable compound because of chlorophyll gradually to fade come to colorless. (74) The field test was examined under the environment and undergoes sunlight. By the reason of the bio-composite film has an easier chance to pale with the time pass, which the color tone of the bio-composite film has fade may affect the UV-light transmittance.



**Table 27 The different color of the film after field test**

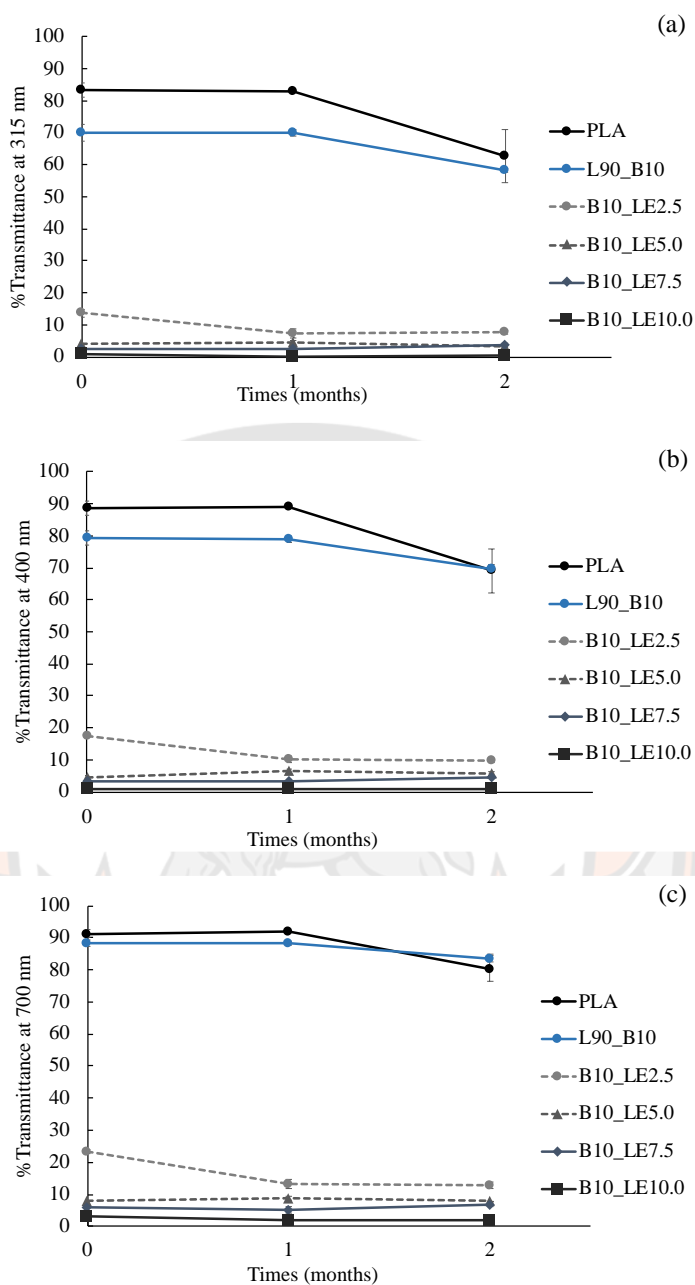
Samples	$\Delta E$ after field test		
	Start	1 months	2 months
Neat PLA film	 Ref.	 1.24	 1.32
L90_B10	 1.20	 0.77	 1.11
B10_LE2.5	 9.73	 4.37	 2.84
B10_LE5.0	 17.13	 9.55	 6.65
B10_LE7.5	 20.48	 10.56	 5.80
B10_LE10.0	 22.29	 13.25	 8.53



**Figure 56** The color parameter (a)  $L^*$ , (b)  $a^*$ , and (c)  $b^*$  values of the film after field test

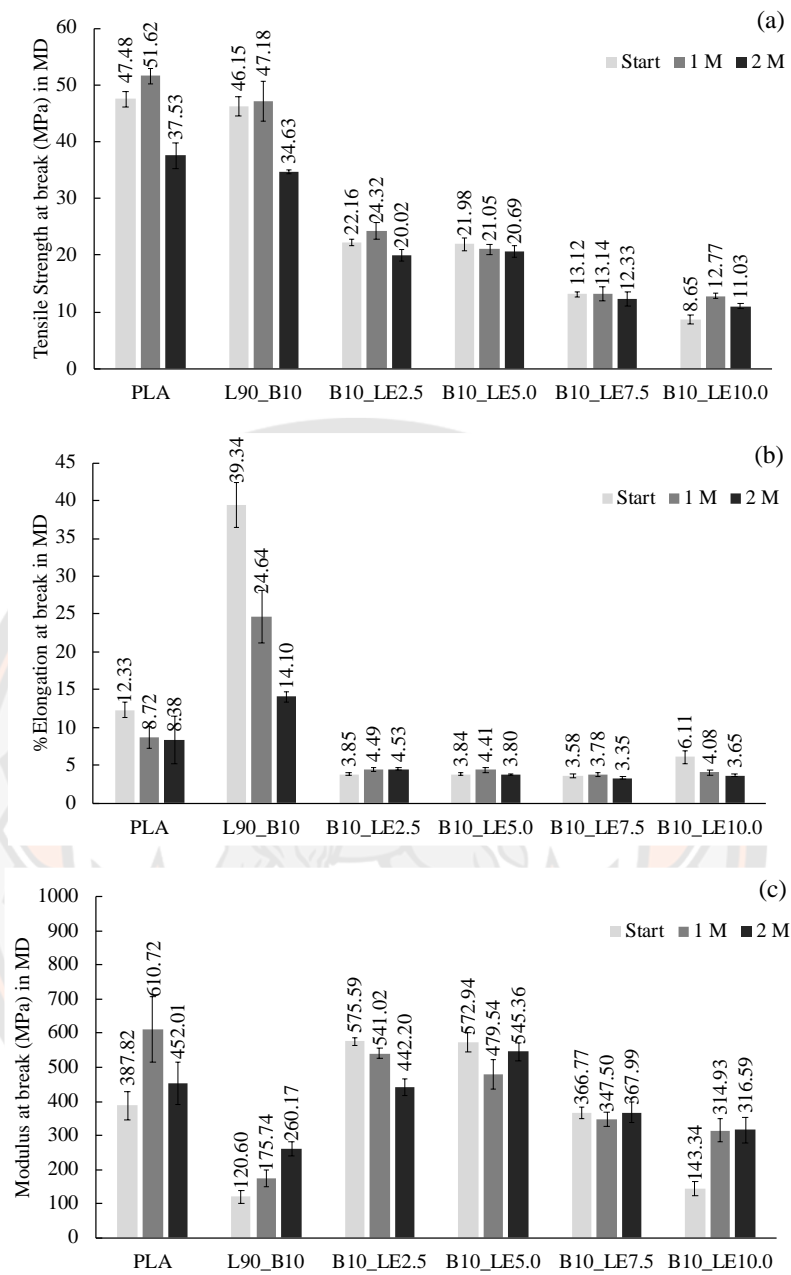
The shade of color has chance to pale of the bio-composite film and slightly affect the UV-light transmission. Figure 57 exhibited the %UV transmittance (%T) of the whole sample after field test. The neat PLA film also has the highest UV-light transmittance. Which the bio-polymer blend film has %T value similar to the neat PLA film. After 2 months, %UV transmittance was decreased due to PLA and PBS can be degraded by photodegradation. While in degradation step, UV radiation reacts within the PLA and PBS polymer chain causing the chemical structure are changed that affects to the polymer chain can be absorbed UV radiation. (35) The bio-composite film also good absorb the UV-light which has the low of %T. The bio-composite film with LE powder also has a good protect the UV-light which the time pass was found %T has value nearly with the film before the field test. Bio-composite film with the highest content of LE powder can absorb the UV-light to more than 90% and the UV-light can transmit through the film around 1-2%.

The %UV-transmittance result of the bio-composite film after field test can explain the addition of the LE powder was reduce the UV-light transmit throughout the bio-composite film. Adding LE powder into the bio-composite film was observed. It was found the LE powder act as a light barrier that can protect the UV-light transmittance. Moreover, the increasing time to field test has hardly effects on the UV-light transmittance. The bio-composite film also has the protect UV-light performance which whole bio-composite film with LE powder has quite a less difference of %UV-transmittance when compare with the start field test was observed in Figure 57.

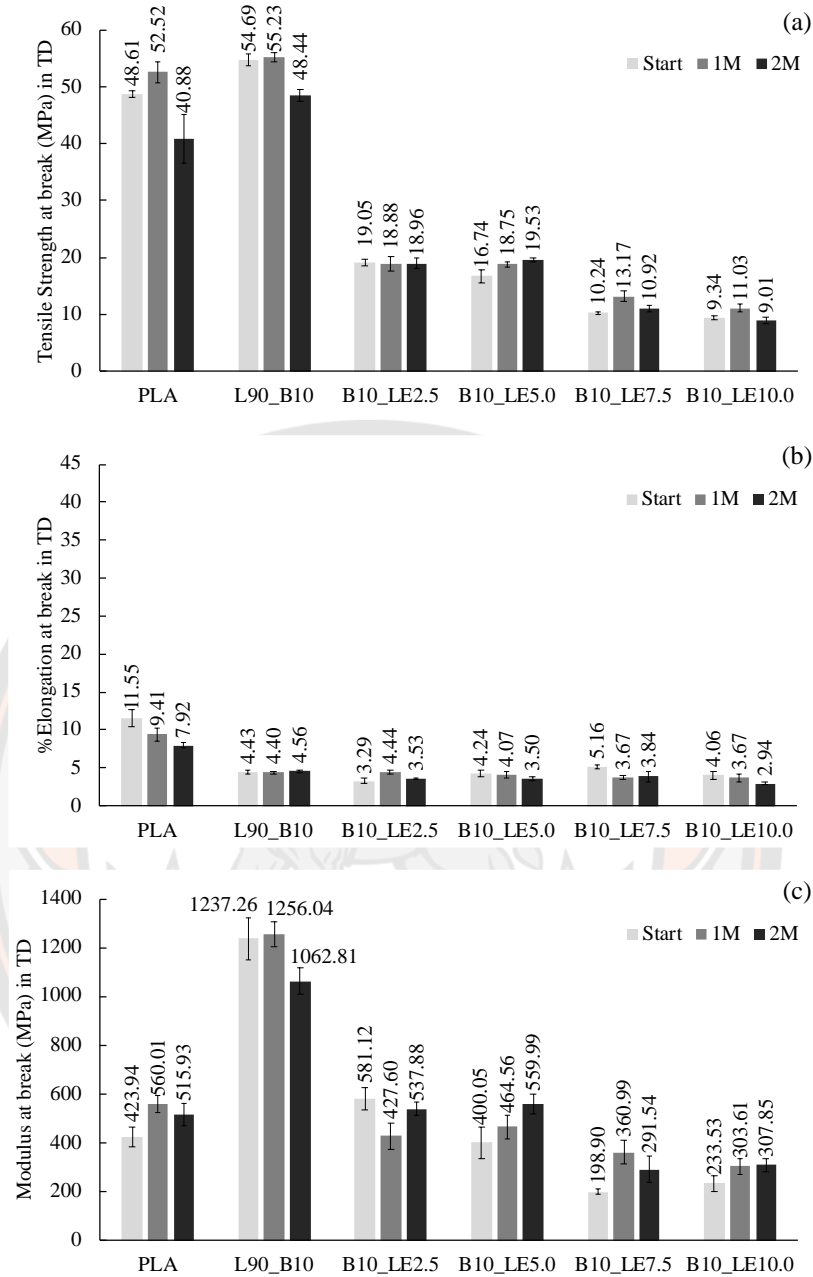


**Figure 57** Percentage of UV-transmittance of the neat PLA film, the bio-polymer blend film, and the bio-composite film after field test in wavelength at (a) 315 nm, (b) 400 nm, and (c) 700 nm

The mechanical properties of the neat PLA film, the bio-polymer blend film, and the bio-composite film after field test in 1 and 2 months was reported in Figure 57 -59. The tensile strength at break of the neat PLA film and the bio-polymer blend film has promising decreased when increasing the time for the field test. While the bio-composite film after field test has the promising to decrease value. However, the value of all the bio-composite films has not quite a different value when compared to before testing. The time pass was found the tendency of %elongation at break of the neat PLA film and the bio-polymer blend film was decreased. While the bio-composite film also has a slight decrease but the value not different from the sample before testing. That may conclude the LE powder act as UV protection. Moreover, the LE powder helps to improve the stability of the bio-composite film. Trend of the tensile strength and %elongation was slightly decreasing and have affected the modulus at break of the whole film when increased. Thus, when time passed causing the film to come to the hard and brittle properties and easier to crack. This result was related to the properties of the main matrix as PLA has hard and brittle properties. (7) The addition of PBS content may improve the mechanical properties of the bio-composite film causing the bio-composite film also to have stretching and flexibility. In conclude, increasing the time of testing has the chance affect the mechanical properties of the bio-composite film was promising to decrease.



**Figure 58** The mechanical properties of all film after field test in machine direction as (a) Tensile strength at break, (b) %elongation at break, and (c) Modulus at break



**Figure 59** The mechanical properties of all film after field test in transverse direction as (a) Tensile strength at break, (b) %elongation at break, and (c) Modulus at break

## CHAPTER V

### CONCLUSIONS

#### Conclusions

This research has been interested in preparing and characterization the mulch film from the bio-composite film based on bio-polymer blend incorporated with the natural fillers. The purpose was to study the effect of natural filler on the physical properties, mechanical properties, thermal properties, biodegradability. The bio-polymer blend used in this work consists of poly(lactic acid); PLA and polybutylene succinate; PBS. The natural fillers were used in this research including ginger that can be separate into 2 stages are early immature ginger (EG) and mature ginger (MG) and another one is lemongrass leaf (LE). The first part was to study and choose the natural filler for appropriate condition to improve the properties of the bio-composite film. Then was to improve the flexibility of the bio-composite film by adding content of PBS. The lase part was to produce and develop the mulch film by produce by choosing the suitable ratio of the bio-polymer blend between PLA and PBS. Which the suitable ratio of bio-polymer blend was incorporated with various content LE powder as 0, 2.5, 5.0, 7.5, and 10.0%w/w, respectively. The mulch film manufactory was prepared in 2 steps, first using a twin-screw extruder were compounding and blow film by a blow film extruder. The film was blown with 70-80 micron as thickness and width with 16-17 cm.

The whole natural fillers have a size distribution with a diameter of approximately 10-45 microns. EG and MG have rather smooth surfaces and non-pore on the surface. The EG and MG particles had attributed to flake and an almost flat circle. LE particle has a rough surface and the shape of particle rather various shape. Which the LE particles had flake, rod shape, an irregular shape. The bio-composite film from the first part was found the bio-composite film incorporated with EG and MG powder had a rougher surface. Due to the EG and MG particles had poor distribution and easier to agglomerate in the bio-composite film. While the LE powder had good distributed in the polymer matrix. The addition of natural fillers affected the mechanical properties that had promising to decrease. The whole bio-



composite film incorporated with each natural filler had a similar trend. The addition of natural fillers can improve the colorlessness of the neat PLA film. The bio-composite film with EG and MG powder had a yellow-brown shade. When incorporated with LE powder was appeared a green-yellow shade. The color of bio-composite film can be improved the UV transmission because the chemical compound of the natural fillers can absorb the UV-light. Which the ginger has curcumin as pigment and the lemongrass leaf has chlorophyll as the pigment. The bio-composite film with LE powder has the highest absorb the UV-light more than 90%. Due to prevention of grass and weed growth in the plot when cover by the bio-composite film. Moreover, the addition of natural fillers can improve the biodegradability of the bio-composite film, which the bio-composite film was fragmentation and after 9 months the bio-composite film had fast degradation.

The flexibility improvement by adding content of PBS. The addition of PBS was various on 10 and 20%w/w. The morphology of the bio-composite film with 10%w/w of PBS (L90\_B10) had smooth surface more than adding to 20%w/w. The addition of PBS had an influence on to increase of the amorphous regions in the bio-polymer blend film. The mechanical properties were found the addition content of PBS at 10%w/w can improve the flexibility of the PLA. The addition of 10%w/w of PBS causing increased %elongation at break from 11-12% to 39%. But adding more content of PBS causing %elongation at break was reducing. The miscibility of the bio-polymer blend film can be considered from the SEM images and the thermal properties. Adding content PBS more than 10%w/w was found Tg and Tm was the obvious separate peak. Moreover, the film appearance of the L80\_B20 had a slightly wrinkle. So, the additional content of PBS at 10%w/w can be improved the flexibility of the PLA. Which the bio-polymer blend film with 10%w/w of PBS has miscibility. While adding more than 10%w/w of PBS influence the bio-polymer blend film comes to the immiscibility. The flexibility improvement by adding content of PBS. The addition of PBS was various on 10 and 20%w/w. The morphology of the bio-composite film with 10%w/w of PBS (L90\_B10) had a smooth surface more than adding to 20%w/w. The addition of PBS had an influence on to increase of the amorphous regions in the bio-polymer blend film. The mechanical properties were found the addition content of PBS at 10%w/w can improve the flexibility of the PLA.

The addition of 10%w/w of PBS causing increased %elongation at break from 11-12% to 39%. But adding more content of PBS causing %elongation at break was reduced. The miscibility of the bio-polymer blend film can consider from the SEM images and the thermal properties. Adding content PBS more than 10%w/w was found T<sub>g</sub> and T<sub>m</sub> was the obvious separate peak. Moreover, the film appearance of the L80\_B20 has a slight wrinkle. So, the additional content of PBS at 10%w/w can improve the flexibility of the PLA. Which the bio-polymer blend film with 10%w/w of PBS has miscibility. While adding more than 10%w/w of PBS influence the bio-polymer blend film comes to the immiscibility. So, from this part was choose the appropriate ratio between PLA and PBS as 90:10. To develop the bio-composite film incorporated with LE powder for applied to produce the mulch film.

The last part was to produce and develop the bio-composite film for using as the mulch film to preventing weed growth. The morphology of the bio-composite film with LE powder had a rough surface and more roughness when increasing the LE powder. The addition of LE powder causing the bio-composite film has amorphous. Adding LE powder to the bio-composite film cause the increasing of crystallinity due to the LE powder help to generate the crystalline of PLA that acts as the nucleating agent. However, the increase of crystalline in the bio-composite film had not increased the mechanical properties. The addition of LE powder to the bio-polymer blend film causing to reduce the mechanical properties. On another hand, the addition of natural fillers can improve the color of the bio-polymer blend film. The bio-composite film had a green-yellow tone. The LE powder can reduce UV-light transmission through the bio-composite film which the UV-light can transit through the bio-composite film just 1-3%. When using the bio-composite film cover on the soil, it can reduce the UV-light transmit to the soil. This was a good performance to protect the damage of the root plant. Moreover, the less %UV-light transmittance to the soil can prevent weed growth. Because the weed must use the UV-light for photosynthesis. When the weed carried the less UV-light causing it cannot grow. The field test was found the sunlight affect the color shade of the bio-composite film was pale. Because the chlorophyll can degrade by the sunlight. The pale of the bio-composite film has hardly affected the UV protection performance. The bio-composite film after the field test in 2 months also has a UV-transmission

performance not different from the bio-composite film before testing. The mechanical properties of the bio-composite film also had not different value when compared with the value before testing. The addition of LE powder had a slightly increase in the water vapor transmittance rate (WVTR). But the bio-composite film can remain to keep the moisture. Thus, covering the soil with the bio-composite film with LE powder can also keep the moisture in the soil.

Therefore, the addition of the LE powder as natural filler and pigment to the bio-composite film. It can improve the colorlessness of the bio-polymer blend film. LE particle was distributed in the bio-composite film act as the barrier for protecting the UV-light. Moreover, the production and development of the bio-composite film to produce the mulch film that can produce by plant machine are a twin-screw extruder and the blow film extruder. This method can be easier to up-scale for the manufactory. The bio-composite film incorporated with LE powder can protect weed growth, keep the moisture in the soil, non-release the toxic chemical, and can be biodegraded in the environment caused to reduce the herbicide. Especially, the choosing of the lemongrass leaf powder as natural filler that is the Thai herbal, which can be increasing the value of the local plant. It can make revenue to agriculturists, communities, and the economy in countries.

### **Suggestion**

1. The bio-composite films in this study can be improved mechanical properties for packaging application. The natural filler should be a treatment to remove the hemicellulose, lignin, and other substance. The treatment of natural filler probably was increased the good interaction between natural filler and the matrix that affects to good mechanical properties of the product.

2. For the field test should be increasing the time for testing. Because some plants need to cover the soil for more than 2 months.

## REFERENCES

1. FAO. The future of food and agriculture – Trends and challenges. Rome: FAO website; 2017.
2. William James L. Plastics: Modifying the Microclimate for the Production of Vegetable Crops. HortTechnology horttech. 2005;15(3):477-81.
3. Compostable Films for Agriculture. Paulo Azevedo2013.
4. Briassoulis D, Babou E, Hiskakis M, Scarascia G, Picuno P, Guarde D, et al. Review, mapping and analysis of the agricultural plastic waste generation and consolidation in Europe. Waste Management & Research. 2013;31(12):1262-78.
5. Steinmetz Z, Wollmann C, Schaefer M, Buchmann C, David J, Tröger J, et al. Plastic mulching in agriculture. Trading short-term agronomic benefits for long-term soil degradation? Science of the Total Environment. 2016;550:690-705.
6. Scarascia G, Sica C, Russo G. Plastic materials in European agriculture: Actual use and perspectives. Journal of Agricultural Engineering. 2012;42.
7. สุกัญญา ร. วัสดุชีวภาพ = Biomaterials: สำนักพิมพ์มหาวิทยาลัยนครสวรรค์; 2561.
8. Paziienza P, De Lucia C. For a new plastics economy in agriculture: Policy reflections on the EU strategy from a local perspective. Journal of Cleaner Production. 2020;253:119844.
9. Mulch Films Market - Industry Analysis, Market Size, Share, Trends, Application Analysis, Growth And Forecast 2021 - 2026: INDUSTRY ARC; 2019 [cited 2021 21]. Available from: <https://www.industryarc.com/Research/Mulch-Films-Market-Research-505106>.
10. Kasirajan S, Ngouajio M. Polyethylene and biodegradable mulches for agricultural applications: a review. Agronomy for Sustainable Development. 2012;32(2):501-29.
11. Tarara J. Microclimate Modification with Plastic Mulch. HortScience. 2000;35.
12. WHAT IS THE DIFFERENCE BETWEEN WHITE MULCH FILM, BLACK MULCH FILM, DOUBLE COLOR FILM, AND COLOR FILM: Henan Yinfeng Plastic; 2018 [cited 2021 8]. Available from: <http://www.mulchfilm.net/what-is-the-difference-between-white-mulch-film-black-mulch-film-double-color-film-and-color-film.html>.
13. Coolong T. Mulches for Weed Management Mulches for Weed Management. 2012.
14. Clear mulch film: famunera; 2016 [cited 2021 8]. Available from: <https://www.famunera.com/product/1272/clear-mulch-film>.
15. Silver Black Mulch Film: IRIS POLYMERS INDUSTRIES 2020 [cited 2021 8]. Available from: <https://www.irispolymers.com/portfolio/silver-black-mulch-film/>.
16. Lamont WJ. 3 - Plastic Mulches for the Production of Vegetable Crops. In: Orzolek MD, editor. A Guide to the Manufacture, Performance, and Potential of Plastics in Agriculture: Elsevier; 2017. p. 45-60.
17. Meckley J. Plastic Film Production. A Guide to the Manufacture, Performance, and Potential of Plastics in Agriculture: s.l. : Elsevier Science; 2017. p. 21-43.
18. Processing and finishing of polymeric materials: Wiley; 2011.
19. Greene JP. Sustainable plastics : environmental assessments of biobased, biodegradable, and recycled plastics: Wiley; 2014.
20. Screw and Check Ring: injectionmoldingonline.com; 2007 [cited 2021 8].

Available from:

<http://injectionmoldingonline.com/Molding101/ScrewAndCheckRing.aspx#>.

21. Shrivastava A. 1 - Introduction to Plastics Engineering. In: Shrivastava A, editor. Introduction to Plastics Engineering: William Andrew Publishing; 2018. p. 1-16.
22. กระบวนการผลิต : การหลอมอัดรีด (EXTRUSION): PTIWISSEN – The Rubber Knowledge; 2019 [cited 2021 8]. Available from: <https://ptiwissen.com/>.
23. Advantages of Co-Rotating Twin Screw Extruder Solution if Compared to Counter-Rotating One: Advanced Compounding and Extrusion systems; [cited 2021 8]. Available from: <https://www.puconsulting.se/wp-content/uploads/2017/04/Co-rotating-VS-counter-rotating-NMC.pdf>.
24. McKeen LW. 3 - Production of Films, Containers, and Membranes. In: McKeen LW, editor. Permeability Properties of Plastics and Elastomers (Third Edition). Oxford: William Andrew Publishing; 2012. p. 39-58.
25. Bhadra J, Alkareem A, Al-Thani N. A review of advances in the preparation and application of polyaniline based thermoset blends and composites. Journal of Polymer Research. 2020;27(5):122.
26. Pracella M. 7 - Blends and Alloys. In: Jasso-Gastinel CF, Kenny JM, editors. Modification of Polymer Properties: William Andrew Publishing; 2017. p. 155-84.
27. Work WJ, Horie K, Hess M, Stepto RFT. Definition of terms related to polymer blends, composites, and multiphase polymeric materials (IUPAC Recommendations 2004). Pure and Applied Chemistry. 2004;76(11):1985-2007.
28. Cholleti E, Gibson I. ABS Nano Composite Materials in Additive Manufacturing. IOP Conference Series: Materials Science and Engineering. 2018;455:012038.
29. ทวีชัย อ. กอมพอลิทีฟเสริมแรงด้วยเส้นใยสั้น: ไอซี ออล ดิจิตอลพรีนซ์; 2559.
30. Carvalho LH, Canedo EL, Farias Neto SR, de Lima AGB, Silva CJ. Moisture Transport Process in Vegetable Fiber Composites: Theory and Analysis for Technological Applications. Industrial & Technological Applications of Transport in Porous Materials. 2013:37-62.
31. Fink JK. The chemistry of bio-based polymers: Scrivener Publishing; 2014.
32. Auras R, Harte B, Selke S. An Overview of Polylactides as Packaging Materials. Macromolecular Bioscience. 2004;4(9):835-64.
33. Zaaba NF, Jaafar M. A review on degradation mechanisms of polylactic acid: Hydrolytic, photodegradative, microbial, and enzymatic degradation. Polymer Engineering & Science. 2020;60(9):2061-75.
34. Hammajam AA, El-Jumrah AM, Ismarrubie ZN. The Green Composites: Millet Husk Fiber (MHF) Filled Poly Lactic Acid (PLA) and Degradability Effects on Environment. Open Journal of Composite Materials. 2019;09(03):300-11.
35. Niaounakis M. 2 - Properties. In: Niaounakis M, editor. Biopolymers: Applications and Trends. Oxford: William Andrew Publishing; 2015. p. 91-138.
36. Suaduang N, Ross S, Ross GM, Pratumshat S, Mahasaranon S. Effect of spent coffee grounds filler on the physical and mechanical properties of poly(lactic acid) bio-composite films. Materials Today: Proceedings. 2019;17:2104-10.
37. Sung SH, Chang Y, Han J. Development of polylactic acid nanocomposite films reinforced with cellulose nanocrystals derived from coffee silverskin. Carbohydrate Polymers. 2017;169:495-503.
38. Lee CW, Kimura Y, Chung J-D. Mechanism of enzymatic degradation of

- poly(butylene succinate). *Macromolecular Research*. 2008;16(7):651-8.
39. Hongsriphan N, Pinpueng A. Properties of Agricultural Films Prepared from Biodegradable Poly(Butylene Succinate) Adding Natural Sorbent and Fertilizer. *Journal of Polymers and the Environment*. 2019;27(2):434-43.
  40. Bhatia A, Gupta R, Bhattacharya S, Choi H. Compatibility of Biodegradable Poly (lactic acid) (PLA) and Poly (butylene succinate) (PBS) Blends for Packaging Application. *Korea Australia Rheology Journal*. 2007;19.
  41. Xanthos M. *Functional fillers for plastics*: Wiley-VCH Verlag; 2005.
  42. Teferra T, Kifle A, Tefera A, Abate S, Sima B, Astatike A, et al. *Ginger (Zingiber Officinale Rosec.): Production, Postharvest Handling, Processing and Marketing - A Comprehensive Extension Package Manual* 2015.
  43. Prasad S, Tyagi A. *Ginger and Its Constituents: Role in Prevention and Treatment of Gastrointestinal Cancer*. *Gastroenterology research and practice*. 2015;2015:142979.
  44. Iijima Y, Joh A. Pigment Composition Responsible for the Pale Yellow Color of Ginger (*Zingiber officinale*) Rhizomes. *Food Science and Technology Research*. 2014;20(5):971-8.
  45. Liu Y, Liu J, Zhang Y. Research Progress on Chemical Constituents of *Zingiber officinale* Roscoe. *BioMed Research International*. 2019;2019:5370823.
  46. ชิง ประโยชน์ดี ๆ สรรพคุณเด่นๆ และข้อมูลงานวิจัย: **DISTHAI**; [cited 2020 20]. Available from: <https://www.disthai.com/16488302/%E0%B8%82%E0%B8%B4%E0%B8%87>.
  47. Chiewchanchairat K, Bumroongsri P, Kheawhom S. *KKU Engineering Journal*. *KKU Engineering Journal*. 2013;40(March):131-8.
  48. Jacob J, Peter G, Thomas S, Haponiuk JT, Gopi S. Chitosan and polyvinyl alcohol nanocomposites with cellulose nanofibers from ginger rhizomes and its antimicrobial activities. *International Journal of Biological Macromolecules*. 2019;129:370-6.
  49. Hashim MA, Yahya F. Effect of different drying methods on the morphological structure, colour profile and citral concentration of Lemongrass (*Cymbopogon citratus*) powder. 2019;7:93-102.
  50. Khonsung P. ตะไคร้. *Thai J Pharmacol*. 2012;34(2):37-51.
  51. Wifek M, Saeed A, Rehman R, Nisar S. Lemongrass: a review on its botany, properties, applications and active components. 2016:79-84.
  52. Alzobaay DA, Kadhim B. Phytochemical Screening, Chemical Composition and Antibacterial Activity of Lemongrass (*Cymbopogon citratus*) Leaves Extracts. 2019.
  53. Ganjewala D, Gupta A. 8 Lemongrass (*Cymbopogon flexuosus* Steud.) Wats Essential Oil: Overview and Biological Activities. 2013.
  54. Bekele LD, Zhang W, Liu Y, Duns GJ, Yu C, Jin L, et al. Preparation and characterization of lemongrass fiber (*Cymbopogon* species) for reinforcing application in thermoplastic composites. *BioResources*. 2017;12(3):5664-81.
  55. Thorat PP, Sawate AR, Patil BM, Kshirsagar RB. Studies on chlorophyll content and colour characteristics of lemongrass (*Cymbopogon citratus*) powder. *International Journal of Chemical Studies*. 2018;6(2):437-9.
  56. Garside P, Wyeth P. Identification of Cellulosic Fibres by FTIR Spectroscopy: Thread and Single Fibre Analysis by Attenuated Total Reflectance. *Studies in Conservation*. 2003;48(4):269-75.
  57. Eyupoglu S. Characterization of New Cellulosic Fibers Obtained from *Zingiber*

Officinale. *Journal of Natural Fibers*. 2020:1-10.

58. Ranade S, Thiagarajan P. Lemon grass. 2015;35:162-7.

59. Sanchez-Garcia MD, Gimenez E, Lagaron JM. Morphology and barrier properties of solvent cast composites of thermoplastic biopolymers and purified cellulose fibers. *Carbohydrate Polymers*. 2008;71(2):235-44.

60. Sengupta D, Mondal B, Mukherjee K. Visible light absorption and photosensitizing properties of spinach leaves and beetroot extracted natural dyes. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*. 2015;148:85-92.

61. Chlorophylls and Carotenoids. 2021.

62. Herzfeld KF. Theory of Light Absorption in Simple Aromatic Compounds. *Proceedings of the American Philosophical Society*. 1940;82(3):359-67.

63. Hartatie E, Prihartini I, Widodo W, Wahyudi A. Bioactive Compounds of Lemongrass (*Cymbopogon citratus*) essential oil from different parts of the plant and distillation methods as natural antioxidant in broiler meat. *IOP Conference Series: Materials Science and Engineering*. 2019;532:012018.

64. Sarath CC, Shanks RA, Thomas S. Chapter 1 - Polymer Blends. In: Thomas S, Shanks R, Chandrasekharakurup S, editors. *Nanostructured Polymer Blends*. Oxford: William Andrew Publishing; 2014. p. 1-14.

65. Veang-in O, Srithep Y. Characterization of Polymer Composites between Poly (lactic acid) and Poly (butylene succinate) with Chain Extender. 2017.

66. Guide to Infrared Spectroscopy: BRUKER; 2021 [cited 2021 25]. Available from: <https://www.bruker.com/content/bruker/int/en/products-and-solutions/infrared-and-raman/ft-ir-routine-spectrometer/what-is-ft-ir-spectroscopy.html>.

67. Olopade BK, Oranusi S, Nwinyi OC, Njobeh PB, Lawal IA. Characterization of Nanoformulations from Montmorillonite clay for the decontamination of zearalenone in cereals using X-ray Diffraction Technique. *Journal of Physics: Conference Series*. 2019;1299(1).

68. Nur Firdaus MY, Osman H, Metselaar HSC, Rozyanty AR. A simple method for the production of pure crystalline silica from lemon grass. *BioResources*. 2015;11(1):1270-9.

69. Mohamed Yusof N, O H, Metselaar H, Rozyanty AR. Preparation and Characterization of Active SiO<sub>2</sub> from *Cymbopogon citratus* Ash Calcined at Different Temperature. *BioResources*. 2016;11.

70. Bouafif H, Koubaa A, Perre P, Cloutier A, Riedl B. Wood Particle/High-Density Polyethylene Composites: Thermal Sensitivity and Nucleating Ability of Wood Particles. *Journal of Applied Polymer Science*. 2009;113:593-600.

71. Španić N, Jambreković V, Šernek M, Medved S. Influence of Natural Fillers on Thermal and Mechanical Properties and Surface Morphology of Cellulose Acetate-Based Biocomposites. *International Journal of Polymer Science*. 2019;2019:1065024.

72. Su S, Kopitzky R, Tolga S, Kabasci S. Polylactide (PLA) and Its Blends with Poly(butylene succinate) (PBS): A Brief Review. *Polymers*. 2019;11(7).

73. Steer J. Structure and Reactions of Chlorophyll [cited 2021 13]. Available from: <https://www.ch.ic.ac.uk/local/projects/steer/>.

74. Ngo T, Zhao Y. Retaining Green Pigments on Thermally Processed Peels-on Green Pears. *Journal of Food Science*. 2005;70(9):C568-C74.