UNIVERSITI PUTRA MALAYSIA

UTILIZATION OF BANANA PSEUDO-STEM WASTE FOR THE PRODUCTION OF FOOD PACKAGING MATERIAL

NURUL ATIQAH NAJHAH BINTI TARMITI

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ABSTRACT

Banana residue is a waste from banana plantation and has the potential to be converted into providing profitable products. Among the residue, banana pseudo-stem is a promising cellulose source that remains underutilized whereby the cellulose from the pseudo-stem has a good potential to be used in a bio-plastic production. To date, there is abundance of waste from the usage of petroleum based plastic thus causing the environmental pollution in the world. Most of the plastic is non-recyclable and non-degradable. Hence, this work was directed towards developing biodegradable films from natural resources such as tapioca starch which is more sustainable and environmentally friendly compared with the petroleum derived plastic.

However, the usage of tapioca starch has been limited due to the poor mechanical and barrier properties. Due to the poor performance of biodegradable starch film, different concentration of banana pseudo-stem powder was incorporated into the biopolymer starch matrix produce starch/banana pseudo stem (BP) films to enhance the properties of the films. Casting method was used to produce the film due to low processing cost and easy fabrication.

The effects of BP concentration on the morphology, optical (color and transparency), mechanical (tensile strength (TS), Young's Modulus (YM) and elongation at break (EAB)) and barrier (water vapor permeability (WVP) and oxygen permeability (OP)) properties of the films were investigated. Although, the optical and mechanical properties of the films reduced when BP concentration was increased, barrier properties of the films improved with the addition of banana pseudo-stem powder on the starch-based film. From the results obtained, it was found that tapioca starch film incorporated with 40% banana pseudo-stem powder was the best composition in order to utilize the BP waste. As a conclusion, it was found that the starch-based films incorporated with banana pseudo-stem powder produced in this study has the great potential to be used in food packaging film production.

ABSTRAK

Sisa pisang adalah sisa dari ladang pisang dan berpotensi untuk dijadikan produk yang menguntungkan. Antara bahan buangan tersebut, pseudo-batang pisang adalah sumber selulosa yang menjanjikan yang masih kurang digunakan di mana selulosa dari pseudo-batang pisang berpotensi besar untuk dijadikan sebagai bioplastik. Sehingga kini, terdapat banyak sisa dari penggunaan plastik berasaskan petroleum yang menyebabkan pencemaran alam sekitar di dunia. Kebanyakan plastik tidak boleh dikitar semula dan tidak terbiodegradasi. Oleh itu, kerja ini untuk menjadikan filem biodegradasi dari sumber semula jadi seperti kanji ubi kayu yang lebih mampan dan mesra alam berbanding dengan plastik petroleum yang diperolehi.

Walau bagaimanapun, penggunaan kanji ubi kayu adalah terhad kerana sifat mekanikal, haba dan halangan yang kurang memuaskan. Oleh kerana prestasi yang kurang memuaskan dari filem kanji terbiodegrasikan, selulosa dimasukkan ke dalam matriks kanji biopolimer untuk meningkatkan sifat filem. Serbuk pseudo batang pisang pada berbeza kepekatan telah dimasukkan ke dalam filem berasaskan kanji untuk membentuk komposit bagi meningkatkan sifat-sifat yang kurang memuaskan ini. Kaedah pemutus digunakan untuk menghasilkan filem kerana kos pemprosesan yang rendah dan senang difabrikasi

Kesan filem kanji/BP pada sifat-sifat morfologi, optik (warna dan ketelusan), mekanikal (kekuatan tegangan (TS), Young's Modulus (YM) dan pemanjangan pada takat putus (EAB)) dan halangan (kebolehtelapan wap air (WVP) dan kebolehtelapan oksigen (OP)) filem telah dikaji. Dari hasil yang diperoleh, didapati bahawa kanji ubi kayu yang digabungkan dengan 40% serbuk pseudo batang banana adalah komposisi terbaik kerana sifatnya dalam mengurangkan sisa. Ciri-ciri warna dan ketelusan filem dikurangkan apabila kepekatan BP meningkat. Ciri-ciri penghalang filem juga bertambah baik dengan menambah serbuk pseudo batang pisang pada filem berasaskan kanji. Sebagai kesimpulan, didapati bahawa filem berasaskan kanji yang dimasukkan dengan serbuk pseudo batang pisang yang dihasilkan dalam kajian ini berpotensi untuk digunakan sebagai filem pembungkusan makanan.

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LIST OF ABBREVIATION

BP	banana pseudo-stem powder
TS	tensile strength
YM	Young's Modulus
EAB	elongation at break
WVTR	water vapor transmission rate
WVP	water vapor permeability
OTR	oxygen transmission rate
OP	oxygen permeability
%	percent
mm	millimeter
ml	milliliter
wt	weight
g	gram
sec	second
°C	degree Celcius
MPa	Mega Pascal
kN	kilo Newton
kV	kilo Volt
RH	relative humidity
SEM	Scanning Electron Microscope
EDX	Energy Dispersive X-tray

L^*	lightness intensity
a^*	redness (a^*)
b^*	yellowness
ΔE^*	total color difference
σ	tensile stress
ϵ	tensile strain

CHAPTER 1

INTRODUCTION

1.1 Overview

Generally, in Malaysia, banana crops produce large amount of residues. Each plant produces only one bunch of bananas and each banana plant cannot be used for the next harvest. After harvesting, the bare pseudo-stems are cut and usually left in the soil plantation to be used as organic material and cause environmental pollution (Cordeiro *et al.*, 2013). Pseudo-stem forms a major waste material in the large scale and this disposal has become a huge problem. It could be estimated that few tons per hectare of banana waste are produced annually (Cordeiro *et al.*, 2013). The pseudo-stem can be profitably utilized for numerous applications and preparation of various products. In this study, banana pseudo-stem powder was utilized to produce the food packaging films due to the edibility, biocompatibility, non-toxicity, non-polluting and low cost. Banana pseudo-stem is fibrous in nature and enriched in cellulose content (Pappu *et al.*, 2014). The major chemical compositions in pseudo-stem are cellulose (55.48%), hemicellulose

(5.35%), lignin (22.25%), pectin (5.65%), wax (1.44%) and extractives (7.59%) (Pappu *et al.*, 2014). Cellulose is usually found in the form of microfibrils, constituted of amorphous and crystalline domains in plant cell wall (Montero *et al.*, 2017).

Packaging is one of the ways to prepare goods for transportation, distribution, storage, retailing and end use (Coles *et al.*, 2010; Al-Naamani *et al.*, 2016). The quality of food product has the potential to deteriorate and contaminate before consumed by the consumer. This is due to the changes in biological, chemical and physical of food products over the time. Packaging prevents quality deterioration, for easier distribution and enhances marketing productivity. Thus, food packaging can extend the shelf life, maintain the quality of food and ensure the safety of food products. Food packaging material is defined as the barrier that protects the food from dirt, moisture, light, air, bacteria and aroma, thus extend the shelf life of food products while maintaining the quality of the food (Quintavalla *et al.*, 2002; Yousuf and Srivastava, 2015). Year by year, technology of food packaging improves and most development in the field of food processing has been oriented towards improving food quality and product. Food packaging technology and improvement are convenient at less cost with higher quality and safety level directly.

Food packaging facilitates storage, handling, transport and preservation of food and is essential to prevent food waste. Besides these beneficial properties, food packaging causes rising concern for the environment due to its high production volume, often short usage time and problems related to waste management and littering. Biodegradable films have the potential to decrease the environmental impact of food packaging. Materials based on renewable resources are being developed at an increasing rate. The only bio-based food packaging materials used commercially on a major scale are based on cellulose. However, materials based on proteins, starch, polylactate and other renewable resources may be the food packaging materials in the future. Additionally, natural biopolymer is susceptible to microorganisms, resulting in good biodegradability, which is one of the most promising aspects of its incorporation in packaging materials and industries.

Recently, people are concern about the environment and demand for an environmentally friendly type of packaging materials. Environmental issues resulted from the used of non-degradable packaging materials such as petroleum-based plastic for packaging is becoming crucial. Conventional plastic is produced from simple hydrocarbon such as ethylene and methane known as an artificial polymer. Synthetic polymer is usually non-degradable, thus lead to the severe environmental problem due to the excessive municipal solid waste on the landfill. Nevertheless, biodegradable packaging material in film form is one of the promising solutions to replace the conventional plastic packaging materials. Biodegradable film is a thin layer of material which can be consumed and provides a barrier for oxygen and solute movement of the food (Bourtoom, 2008; Resa *et al.*, 2012). The benefits of using this type of film include non-toxic, renewable and can reduce the use of petrochemical-based packaging material. However, despite the many good benefits, biodegradable film is still unable to replace synthetic packaging which has better mechanical, barrier and thermal property.

Biodegradable film can be produced from natural biopolymers such as polysaccharides, proteins and lipids (Philips *et al.*, 2004; Martins *et al.*, 2012). For this study, starch, which falls in polysaccharide family, is chosen to produce the films because starch is abundant, cheap and non-toxic. Starch is one of the most promising bio-based packaging materials since starch is a renewable source, easy to handle, low in cost and available in market (Resa *et al.*, 2012). Starch is mainly found in the tuber plant and maize plant. The main components of starch granules consist of linear amylose and highly branched amylopectin that formed by the glucose units via α–1,4 bonds (Chiellini, 2008; Ji *et al.*, 2017). Both amylose and amylopectin exhibit different properties whereby amylose is stronger than amylopectin due to high crystallinity of amylose compared to amylopectin. Thus, the ratio of amylose and amylopectin in the starch may affect the properties of the film.

Although the native starch can be used to produce biodegradable film, however the films exhibit poor mechanical and thermal properties. One of the effective ways to overcome this limitation is by incorporating other biopolymer with the starch film. Generally, the blends of biopolymers with other biopolymers have been widely used to improve the physical, mechanical, functional and nutritional properties of the prepared biodegradable films (Vásconez *et al.*, 2009). Cellulose appears to be a potential biopolymer incorporated with starch due to its superior thermal conductivity and mechanical properties. Cellulose can be added to starch film in order to enhance the mechanical properties and barrier resistance of the film (Medeiros *et al.*, 2010; Ferracane *et al.*, 2014). Cellulose has been proved promising as bio-composite materials in film packaging and shows satisfactory physical properties such as relatively high tensile strength, high stiffness and high Young's Modulus (Zainuddin, 2009).

1.2 Problem statement

In general, banana residue is an abundant natural source in subtropical and tropical regions. After harvesting banana bunches from the trees, a large amount of waste biomass remains because each banana plant cannot be used for the next harvest. Banana pseudo-stem normally felled and usually abandoned in the soil plantation to become organic waste and cause environmental pollution. This can lead to a serious environmental issue, especially for huge scale farming industries if they are not practicing the proper agricultural waste management. However, the banana pseudo-stem has the potential to be converted into profitable products. The banana pseudo-stem is a promising source that remains underutilized. To overcome this problem, the food packaging material is produced from banana pseudo-stem due to the high cellulose content in the stem and its eco-friendly property.

Packaging materials that is biodegradable can be produced from biopolymers such as starch. Developing environmentally friendly packaging materials that is biodegradable and edible is a promising way to reduce landfill waste (Arvanitoyannis and Kasaverti, 2008). Starch is particularly regarded as an ideal raw material for the production of biodegradable polymers due to its ability to form a continuous matrix, low permeability to oxygen in comparison to non-starch films, availability and low cost (Jiménez *et al.*, 2012). Starch films also have been found to be transparent, odorless and colorless which is a good packaging material (Jiménez *et al.*, 2012). However, the usage of the starch film has been limited due to poor mechanical, thermal and barrier properties but this properties can be improve through the addition of other film-forming materials (Duncan, 2011). Nonetheless, concentration of the biopolymer is imperative parameters that need to be determined in order to produce efficient packaging film. Thus, this work is directed to investigate the effect of concentration of banana pseudo-stem powder on the mechanical properties, thermal properties and barrier properties of the starch film.

1.3 Objectives

The general objective of this work was to investigate the properties of starch-based films incorporated with banana pseudo-stem powder. The specific objectives of this work were:

1. To produce and characterize the morphological and optical properties of starch-based films incorporated with different amount of banana pseudo-stem powder.
2. To investigate the mechanical and barrier properties of starch-based films incorporated with different amount of banana pseudo-stem powder.

1.4 Thesis outline

This thesis is divided into five chapters. Chapter 1 begins with the background of banana plant waste, food packaging materials and overview about starch-based film. Chapter 1 also contains the problem statement, objectives and thesis outline. Chapter 2 covers a detailed literature review on the banana pseudo-stem, starch based films, biodegradable food packaging film and as well as their important properties. Chapter 2 also explains the method used to analyze morphology, optical, mechanical and barrier properties of the films. Chapter 3 presents the materials and methodology used to prepare the films as well as materials and method used to determine the morphology, optical, mechanical and barrier properties of the developed films. Chapter 4 presents the results and discussion of the findings obtained throughout the study. Chapter 5 concludes all the results of the work as well as some recommendations for potential future work.

CHAPTER 2

LITERATURE REVIEW

2.1 Banana pseudo-stem

Banana residues such as pseudo-stem are discharged as an agricultural waste that good for soil plantation after harvesting. About 88% mass of the banana plant produces agricultural waste, especially leaf and banana pseudo-stem after harvesting (Elanthikkal *et al.*, 2010; Li *et al.*, 2015). This agricultural waste leads to a serious environmental issue, especially for huge scale farming industries if they are not practicing the proper agricultural waste management. In this study, powder is obtained from banana residue which is banana pseudo-stem as a composite in starch film. Banana pseudo-stem type *Musa acuminata X balbisiana ABB cv. Awak* is used in this study as shown in *Figure 2.1*.

Figure 2.1*: *Musa acuminata X balbisiana ABB cv. Awak

Banana pseudo-stem is a promising cellulose source that remains underutilized. The major chemical compositions in banana pseudo-stems are cellulose and lignin. Lignin content in banana pseudo-stem is less compared to other stem. *Table 2.1* shows the major chemical constituents of banana tree residue from the work done by Bibla *et al.* (2012). The chemical constituent of banana residue depends on the species and also with its growing environmental condition. Cellulose from banana pseudo-stem has a great potential to be formed as a bioplastic (Faradila *et al.*, 2017).

***Table 2.1:* Elemental analysis and components analysis of banana tree residues (Rosal, 2012)**

Parameter (%)	Experimental	Bilba et al. (2007)
Carbon	39.67	36.83
Hydrogen	5.65	5.19
Nitrogen	1.44	0.93
Sulfur	0.05	-
Cellulose	55.48	46.25
Lignin	22.25	15.07
Extractives	7.59	4.46
Ash	15.35	8.65

Cellulose leads to the improvement of film properties and has expanded consideration and interest because of its potential benefit in term of renewability, biodegradability and high surface area for bonding. It is a common polysaccharide resource that is produced by plants and composed of glucose monomers that form a linear polymer with very long macromolecular chains as shown in *Figure 2.2*. Cellulose is highly crystalline, brittle, infusible (Chandra *et al.*, 2007; Ji *et al.*, 2017), lightweight and high-strength particle (Podsiadlo *et al.*, 2005; Ji *et al.*, 2017). The combination of starch and cellulose also can greatly enhance the tensile properties of a resulting composite. For example, poly(styrene-co-butyl acrylate) latex film that contained 30 wt.% of straw cellulose show a modulus higher than bulk because of the great formation of a fibrils network within a polymer matrix (Helbert *et al.*, 1996; Tan *et al.*, 2016).

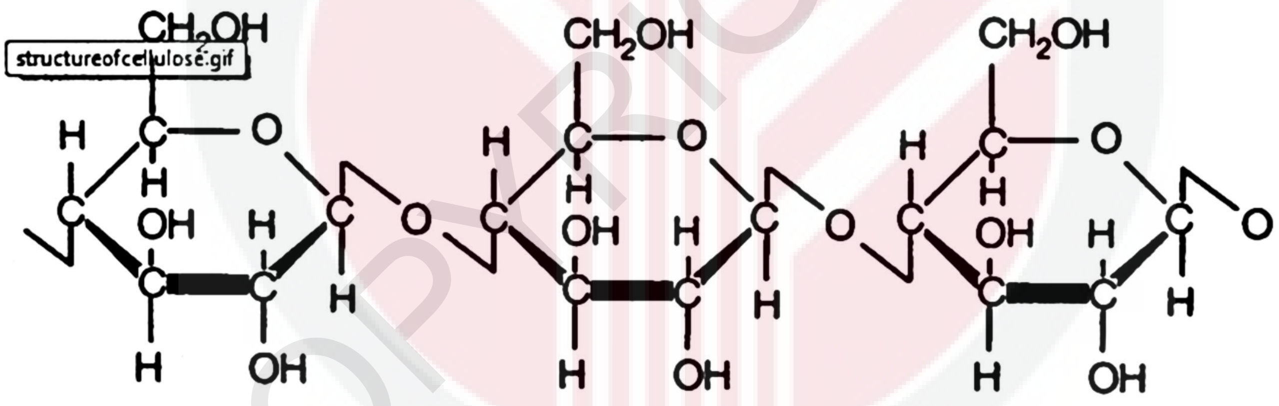


***Figure 2.2*: Structure of cellulose (Chloe Van et al., 2010)**

There are a few of researches that focused on the effect of cellulose incorporated with starch-based film performance. As reported, the combination of cellulose and starch enhances the thermomechanical properties, reduces the water sensitivity and keeps the biodegradability properties of the film (Lima and Borsali, 2004; Xie *et al.*, 2013). The cellulose incorporated with starch also increases the glass transition temperature (T_g) (Alemdar and Sain, 2008; Panaitescu *et al.*, 2016). Network within cellulose and starch resulting the formation of strong hydrogen bonds. The brittleness of starch-based film also decreased and the water uptake by starch-based film decreased with increasing of cellulose reinforcement (Lu *et al.*, 2005; Ferracane *et al.*, 2014).

2.2 Starch-based edible film

Biodegradable film is the most beneficial alternative food packaging material because it is environmentally friendly and has the ability to extend shelf life, maintain the quality of the food and control freshness the product. In this work, biopolymer used is from the first category which is starch from polysaccharide family (Martins *et al.*, 2012). Starch is a natural polymer that can be is found in nature. Starch is a major carbohydrate resources found in a tubular plant eg: cassava, potato, sweet potato) and cereal plant (eg: corn, maize, and rice). In industry, starch is used as a thickener in processed foods and in the manufacture of paper, biodegradable plastics and adhesives. As a subsequence it is easily available and very cheap.

There are many types of starch that can be used to prepare biodegradable film including tapioca starch, sweet potato starch, yam starch and corn starch. In this work, tapioca starch has been chosen as the biopolymer material to prepare biodegradable film due to its availability and relatively low cost (Souza *e al.*, 2012). Tapioca starch has high viscosity, high clarity and medium stability as shown in *Table 2.2* (Gbadeg, 2013). Besides, tapioca is a potential starch material for polymer film packaging as it is isotropic, non-toxic, odorless, tasteless, colorless, flexible and biologically degradable. It also exhibits appropriate physical characteristics and are impermeable to oxygen from the environment in film form.

***Table 2.2*: Properties of tuber crops starches (Gbadeg, 2013)**

TUBERS	VISCOSITY	CLARITY	STABILITY
Tapioca	High	High	Medium
Sweet potato	Medium-high	High	Medium
Yam	Low	High	High
Aroid	Low-medium	Low	High
Canna	High	High	High
Arrowroot	Medium-high	Medium	Medium

Starch contains granules composed of two polymers of glucose which are amylose and amylopectin as shown in *Figure 2.3*. Amylose is responsible for the film-forming capacity of starch based films. Amylopectin contains 2–4% branching points formed by α–1,6 bonds of the main backbone and other branches (Chiellini, 2008; Ji *et al.*, 2017). The amylopectin and amylose ratio in starch may affect the crystallinity of starch based product (Zhang *et al.*, 2017). The crystallinity is proportional to the amylose content whereby high crystallinity will produce firmer structure of film.

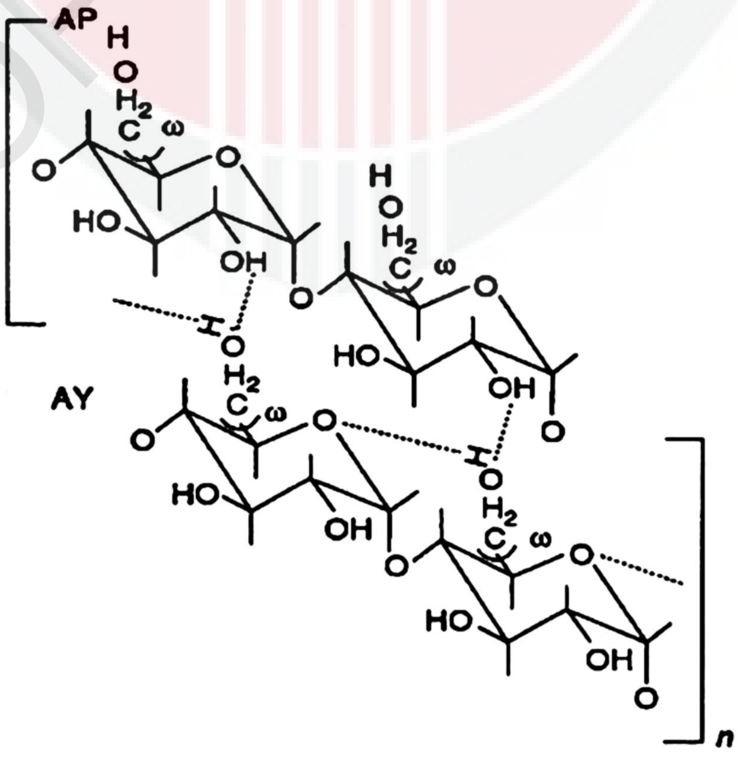


***Figure 2.3*: Hydrogen bonding between amylose and amylopectin molecules in starch (Tako & Hizukuri, 2002)**

Starch granules are insoluble in water at low temperature because the swelling in water is limited at low temperature. At high temperature, it may lead to the ultimate dispersion of most of the granules substance which called gelatinization. During the gelatinization process, starch is heated continuously in excess water. The starch granules swell and the ordered structures of starch molecules are disrupted and thus increase the viscosity of starch solution. Starch gelatinization properties and swelling granules as well as pasting properties have been shown to be affected by amylose content (Ferracane *et al.*, 2014). In the presence of a certain range of temperature and excess water, granules will lose their ordered structure of gelatinization. Viscosity will increase and a process called pasting will be resulted if further heating at temperature higher than gelatinization temperature range. Swelling of the granule is caused by the water adsorption in the amorphous region of the starch granules (Bogracheva *et al.*, 2002; Moura *et al.*, 2015). Amorphous region is free for water adsorption which increases swelling and followed by disruption of ordered structures.

One of the effective ways to overcome the strong limitations of the starch-based films such as poor tensile properties and thermal properties is to strengthen the starch with other biopolymer form composite film (Cyras *et al.*, 2008; Ferracane *et al.*, 2014). This is because the biocomposites exhibit a high level of efficiency in improving the mechanical and thermal properties of starch-based films. These particles also have a good compatibility with the matrix in the thermoplastic starch films. Additional of other biopolymer is expected to withstand the stress of thermal food processing, transportation and storage (Thostenson *et al.*, 2005, Al-Naamani *et al.*, 2016). Polymer incorporated with inorganic polymer such as epoxy, polypropylene, polyvinyl chloride, polyamide, polystyrene and polymethyl methacrylate improved in terms of mechanical and barrier properties.

2.3 Biodegradable food packaging

Food packaging plays an important role in protecting and preserving food during the distribution chain. Nowadays, food packaging is widely used in food industry around the world to control the benefits of food after processing is complete. Packaging also allows foods to travel from their point of origin, ensures safe delivery of the product, hygienic at the time of consumption and provides consumers with ingredient and nutritional information. Without packaging, the food can become contaminated by direct contact with external influences such as physical, chemical and biological contaminants (Yousuf and Srivastava, 2015). Recently, wide range of materials have been used as packaging materials including metal, glass, wood, paper or pulp-based materials, plastics, ceramics or a combination of more than one material as composites (Al-Naamani *et al.*, 2016). Each of the material has different mechanical, thermal, barrier, optical and morphological characterization. The right selection of packaging material is important to maintain the quality of the food and control freshness during distribution and storage.

Biodegradable packaging is usually made from bio-based materials or biopolymer which derived from renewable sources. These materials can be used for food application (Claus, 2000; Yousuf and Srivastava, 2015). The natural biopolymers that are used for food packaging are biocompatible and biodegradable and these characteristics lead to ecological safety (Prashanth *et al.*, 2007; Al-Naamani *et al.*, 2016). The combination of biopolymer with other biopolymer in preparation of films is directly effective on the properties such as thermal stability, flexibility, good barrier to gases, good barrier to water, resistance to chemicals, biocompatibility and biodegradability (Guber *et al.*, 2006; Malho *et al.*, 2015). Biopolymer-based packaging materials originated from naturally renewable resources such as polysaccharides, proteins and lipids. Combinations of those components have the potential to replace the current synthetic plastics (Sothornvit, 2009; Resa *et al.*, 2012).

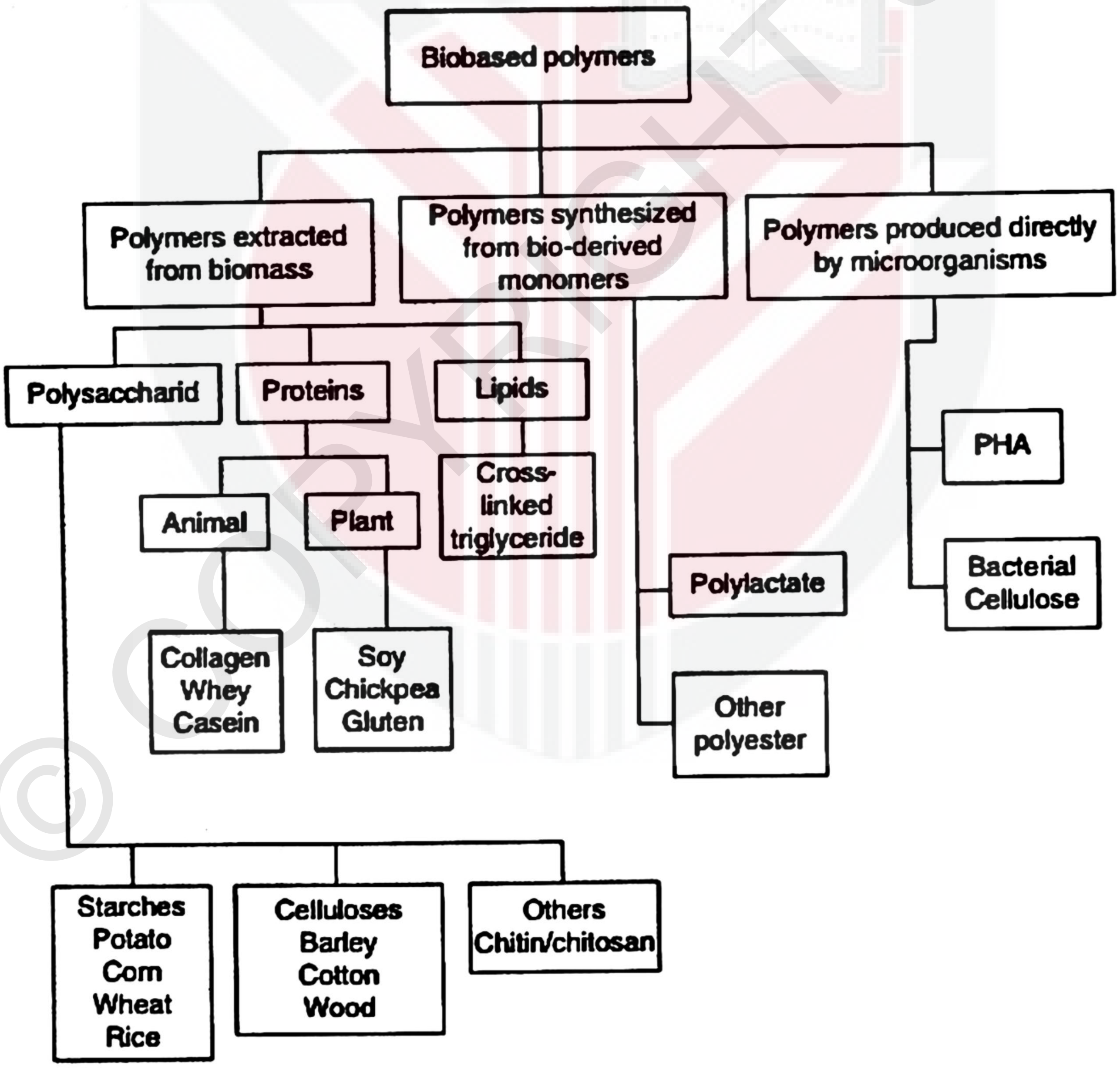


***Figure 2.4*: Bio-based polymers for food packaging (van Tuil *et al.*, 2000)**

Bio-based polymers can be divided into three main categories as shown in *Figure 2.4*. These categories are based on their origin and production. The first category is a polymer directly extracted from biomass. For example polysaccharides such as starch and cellulose and protein like casein and gluten. The second category is a polymer produced by classical synthesis using renewable bio-based monomers such as polylactic acid. These monomers may be produced by fermentation of carbohydrate feedstock. The last one is polymers produced by microorganisms or genetically modified bacteria.

Several concerns must be addressed prior to commercial use of bio-based primary food packaging materials. These concerns include degradation rates under various conditions, changes in mechanical properties during storage, potential for microbial growth and release of harmful compounds into the packaged food product. Furthermore, the bio-packaging must function as food packaging, meet the requirements of the individual food product and evaluates the suitability of bio-based packaging for foods (Petersen *et al.*, 2012). Physical changes include softening, toughening, loss of solubility, loss of water holding capacity, wetting, lumpiness, caking, emulsion instability, swelling, shrinkage and crushing of food are important in the selection of packaging materials.

2.4 Morphological Properties

The morphological structure of polymer composites is an important characteristic because it will ultimately determine the many properties of the polymer composites. Morphologies of the surface of neat starch and starch/BP film have been observed using Scanning Electron Microscope (SEM). This type of equipment uses high-energy beam of electron directed to the sample for the purpose of inspecting morphology of samples (Schweitzer *et al.*, 2014).

2.5 Optical properties

Generally, packaging films should exhibit high gloss and high transparency properties for a good visual presentation of products. The optical properties of the packaging films are important factors in terms of consumer acceptability as packaging material. Starch-based films showed homogeneous surfaces and compact structures (Li *et al.*, 2015). The addition of the banana pseudo-stem powder led to the formation of films with more heterogeneous surfaces where large size particles of the material covered by the starch matrix. The fact that cellulose was coated with the starch matrix indicated that the residue was structurally incorporated in the network, owing this to a high compatibility between both components.

Starch-based films are apparently transparent. The transparency rate is usually decrease after the addition of other biopolymer due to the formation of two phases in the system. The higher the amount of cellulose will decrease the transparency of the films.

Table 2.3 shows the transparent values *L**, *a**, *b** of the starch-based films with and without addition of cellulose (Li *et al.*, 2015). Transparency and color of the film were affected by the percentage of cellulose. Contrast to control film, the composite films were less transparent with the increased in percentage of cellulose (Li *et al.*, 2015) as shown in *Table 2*.3. Film transparency decreased significantly with an increase in concentration in thermoplastic starch formulations and reduced light transmittance of films (Castillo *et al.*, 2013). The lower transparency of films has the ability to protection of package contents from light and enhances the quality of the packaged food.

***Table 2.3*: Color parameters of starch-based films with and without cellulose (Li et al., 2015)**

Sample	*L**	*a**	*b**
Pea starch	86.93 ± 0.15	1.15 ± 0.01	2.10 ± 0.02
Pea starch + 1% cellulose	86.71 ± 0.35	1.11 ± 0.01	2.29 ± 0.03
Pea starch + 3% cellulose	87.39 ± 0.18	1.13 ± 0.04	2.46 ± 0.09
Pea starch + 5% cellulose	86.81 ± 0.05	0.08 ± 0.03	2.55 ± 0.17
Pea starch + 7% cellulose	87.33 ± 0.18	1.05 ± 0.03	3.22 ± 0.10
Pea starch + 9% cellulose	86.79 ± 0.28	1.07 ± 0.06	4.01 ± 0.13

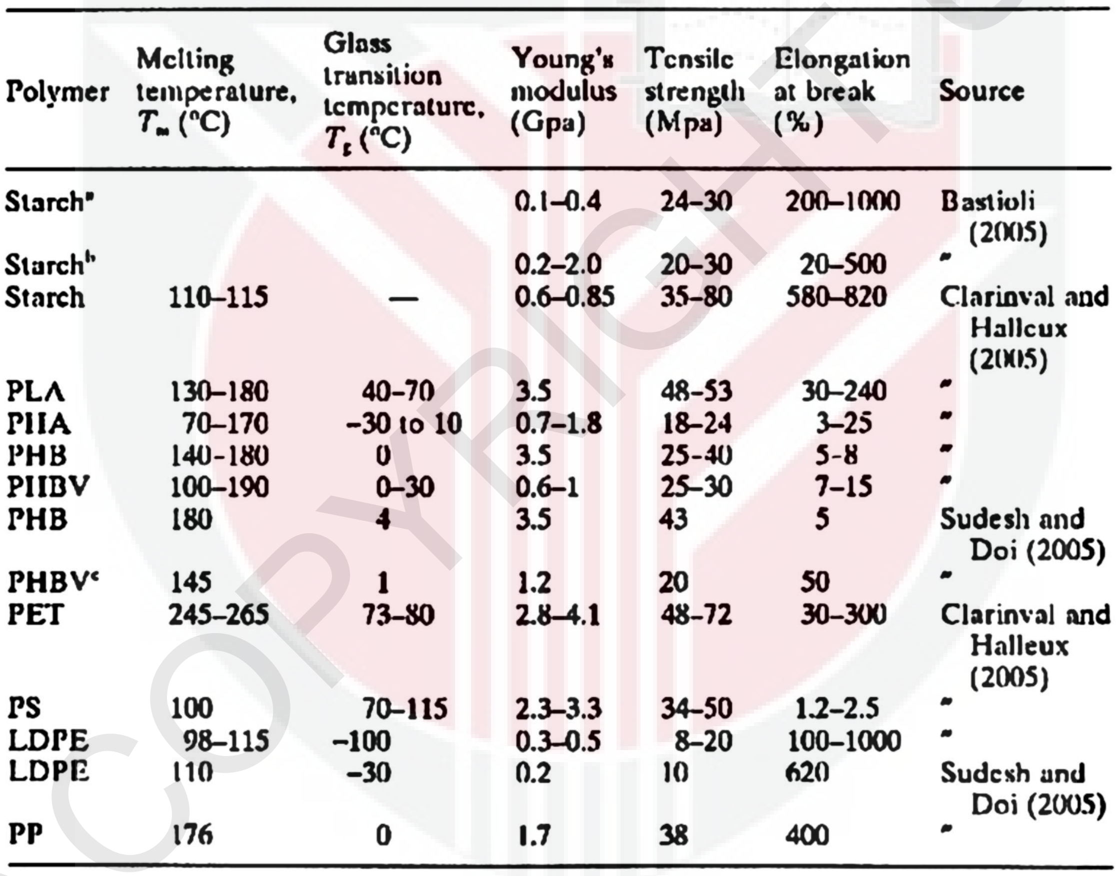
2.6 Mechanical properties

The aim of determining mechanical properties of the films is to determine the tensile strength, elastic modulus and deformation at break of the sample whereby the stress-strain curves are usually acquired and analyzed (Chillo *et al.*, 2008; He *at al.*, 2017). High tensile strength (TS) enables packaging materials to tolerate the typical stress encountered by packaging materials during food handling and transportation (Sadegh-Hassani and Nafchi, 2014). In addition, tensile strength of the material is the maximum amount of tensile stress that a sample can take before break point of the sample (Gerschutz et al., 2011; He at al., 2017). The failure is indicated at the break point of the sample. Besides, tensile strain is the measure of the changes in the length of the sample relative to its original length when the sample is subjected to tensile stress (Hanpin and Stephen, 2013).

Young's Modulus (YM) is the measurement of stiffness of the sample or the force that is required to deform the sample film (Gilfillan et al., 2012). The elasticity of the films is related to interactions of the molecules in the film. The sample has a perfect elasticity if the sample shows an immediate recovery to its original length and size after deformation. Elongation at break (EAB) defines as the change of the sample from its original length in the stress-strain experiment in the break point. It is also known as fracture strain and is the ratio between change in the length of sample film and initial length after breakage of the test specimen (Hanpin and Stephen, 2013).

Mechanical properties of bio-based and petroleum derived polymer are totally different as shown in *Table 2.4*. Bio-based polymers are materials which are produced from renewable resources. Bio-based polymers not only can replace existing polymers in a number of applications but also provide new combinations of properties for new applications. In *Table 2.4,* the high mechanical strength of starch-based films is attributed to extensive attraction hydrogen bonding between film polymer molecules that create a strong cohesive network. Starch-based film has relatively stronger mechanically than protein based film (Rompothi *et al*., 2016).

***Table 2.4*: Mechanical properties of bio-based and petroleum derived polymer (Chiellini, 2008)**



Polymer	Melting temperature, T_m (°C)	Glass transition temperature, T_g (°C)	Young's modulus (Gpa)	Tensile strength (Mpa)	Elongation at break (%)	Source
Starch^a			0.1–0.4	24–30	200–1000	Bastioli (2005)
Starch^b			0.2–2.0	20–30	20–500	"
Starch	110–115	—	0.6–0.85	35–80	580–820	Clarinval and Halleux (2005)
PLA	130–180	40–70	3.5	48–53	30–240	"
PHA	70–170	−30 to 10	0.7–1.8	18–24	3–25	"
PHB	140–180	0	3.5	25–40	5–8	"
PHBV	100–190	0–30	0.6–1	25–30	7–15	"
PHB	180	4	3.5	43	5	Sudesh and Doi (2005)
PHBV^c	145	1	1.2	20	50	"
PET	245–265	73–80	2.8–4.1	48–72	30–300	Clarinval and Halleux (2005)
PS	100	70–115	2.3–3.3	34–50	1.2–2.5	"
LDPE	98–115	−100	0.3–0.5	8–20	100–1000	"
LDPE	110	−30	0.2	10	620	Sudesh and Doi (2005)
PP	176	0	1.7	38	400	"

The combination of starch with other biopolymer exhibits a remarkable improvement in the mechanical properties especially in the Young's Modulus (Cyras *et al*., 2008, He *at al*., 2017). The main reason related in the mechanical properties is the stronger interfacial interaction between the biopolymer. Addition other biopolymer such as cellulose usually effected the mechanical properties of starch-based films (López *et al.,* 2015). Performance of TS of starch films can be improved by produce composite film (Wu *et al.*, 2009; Rompothi *et al.*, 2017). It is likely that incorporation of particles occupy the sites on starch that normally would be occupied by water. Mechanical properties of the composite films have been reported to be highly dependent on the interfacial interaction between the matrix (Yamaoki *et al.*, 2009; Savadekar, 2012).

Incorporated films presented higher values of maximum tensile strength and Young's Modulus and lower elongation at break, if compared to starch-based films without cellulose addition as shown in *Table 2.5*. Starch reportedly produces films with higher TS (Piyada *et al.*, 2013). Increase in TS is of important criteria for food packaging applications as high TS can allow the packaging films to withstand the normal stress encountered during food handling, shipping and transportation. This is because the linear structure chains enhance its ability to form hydrogen bonds in the starch matrix (Piyada *et al.*, 2013). It is related to the interfacial interaction between the biopolymer. The elasticity of the films is related to interactions of the molecules (Hassani and Nafchi, 2014).

***Table 2.5*: Mechanical properties of cellulose and potato starch composite films (Hassani & Nafchi, 2014)**

Cellulose (%)	Tensile strength (MPa)	Elongation at break (%)	Young modulus (MPa)
0	7.33 ± 0.34	68.0 ± 1.8	188 ± 14
1	8.09 ± 0.49	61.5 ± 2.2	297 ± 5
2	8.27 ± 0.32	54.5 ± 6.1	315 ± 7
3	8.45 ± 0.25	52.4 ± 3.5	332 ± 14
5	9.82 ± 0.42	44.0 ± 3.9	376 ± 12

2.7 Barrier properties

Packaging material must be able to protect the food product from moisture, gas and other volatile materials in order to extend shelf life of the food. Several factors that influence the barrier properties of a packaging material include temperature and relative humidity. Permeates diffusion across a film is influenced by the film structure, film permeability to specific gases or vapor, thickness, area, temperature, the difference in pressure or concentration gradient across the film (Mukurumbira *et al.*, 2017). *Figure 2.5* shows the mechanism of gas or vapor permeation through a film. In this study, barrier properties which include water vapor permeability (WVP) and oxygen permeability (OP) were determined. The permeability of water vapor and oxygen through a film are generally regarded as a function of solubility and diffusion (Han, 2014).

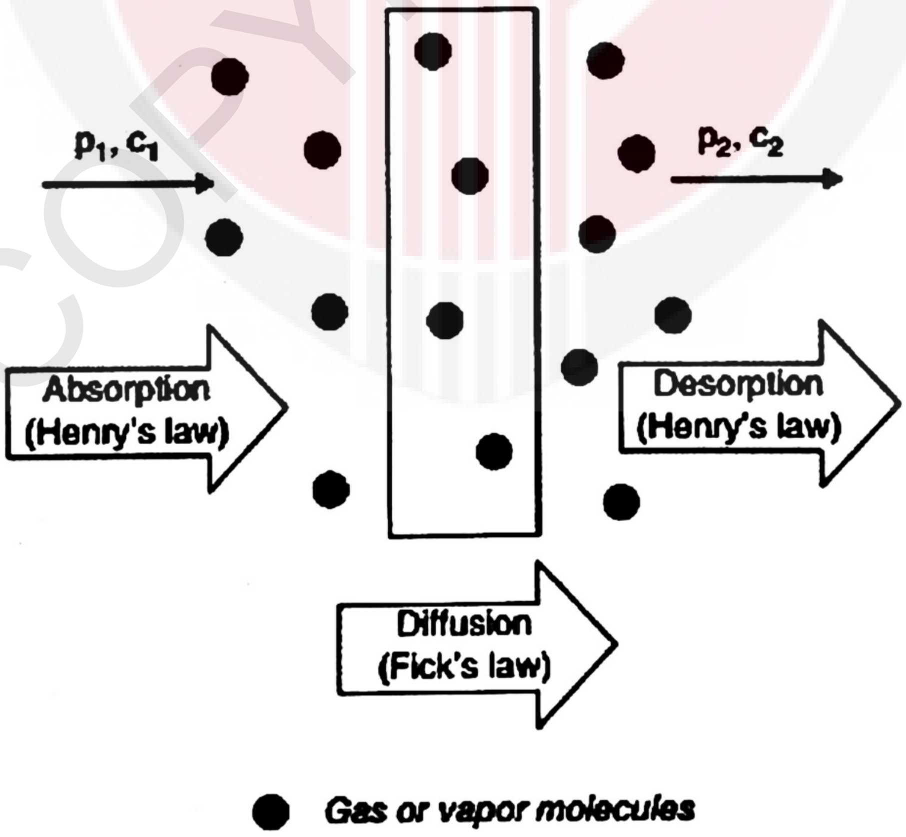


***Figure 2.5*: General mechanism of gas or vapor permeation through a film (Siracusa,2012)**

Water vapor permeability is a measure of the passage of water vapor through the material. Food packaging material must exhibit well to low water vapor permeability to avoid or decrease the moisture transfer between the food and the surrounding atmosphere. In this study, the apparatus and method in American Society for Testing and Materials ASTM E96 (ASTM, 2010) were used to measure the WVP of the film. Water vapor permeability also known as a water vapor transmission rate (WVTR) or moisture vapor transmission rate (MVTR). Previous reports stated that the WVTR of polysaccharide films were related to their thickness (Mylla¨rinen *et al.*, 2002; Xingli *et al.*, 2017) whereby WVTR can be estimated by dividing the slope with the film surface area. WVP can be used to predict the loss or gain of water in the food covered by the film. The different types of starch significantly influenced the WVP of the starch as shown in *Table 2.6*. This might be due to the effect of the different chemical composition especially in starch content.

***Table 2.6*: Comparison of WVP values of edible films (Chiellini, 2008)**

Film formulation	**WVP (g m^{-1} s^{-1} Pa^{-1})**	**Reference**
Yam starch	$(0.96\times1.81)\times10^{-10}$	Mali *et al.* (2002)
Corn starch	$(5.37\times6.70)\times10^{-10}$	Mali *et al.* (2002)
Tapioca starch	$(4.02\times6.25)\times10^{-10}$	Mali *et al.* (2002)
Pea starch	$(7.65\times37.72)\times10^{-10}$	Zhang & Han (2006)
Wheat gluten	$3.8\times10^{-11} - 4.1\times10^{-7}$	Roy *et al.* (2000)
Pea protein	$(1.14\times2.06)\times10^{-9}$	Choi & Han (2001)
Corn zein	$(4.7\times8.9)\times10^{-10}$	Koh *et al.* (2002)

Oxygen barrier can be determined by the oxygen transmission rate (OTR) which indicates the amount of oxygen that permeates per unit of area and time through a packaging material. Oxygen promotes deteriorative reactions to food such as browning reactions, aerobic microbial growth and fat oxidation and thus can shorten the shelf of food (Joongmin *et al.*, 2014). The oxygen pressure inside the container will drop to the point where the oxidation is retarded when the samples have low oxygen permeability coefficients.

Starch has high oxygen permeability but the decrease in oxygen transmission rate (OTR) of composite films due to which generates a more tortuous path for the permeation of oxygen molecules presence of filler dispersed phase in the biopolymer matrix. Oxygen transmission rate reduced when the increasing of the cellulose in the thermoplastic starch film (Savadekar, 2012). Moreover, the presence of other biopolymer appeared to have reduced the affinity of the starch films for water and form the strong reinforcement interface between the matrices. *Table 2.7* shows the effect of addition of composite on the water vapor transmission rate and oxygen permeability through starch-based films which is potato starch (Hassani & Nafchi, 2014).

Addition of other composite significantly decreased WVP and OP of potato films because the particles bind the pores in the molecular structures and can decrease oxygen permeability as well as permeability to water vapor. Müller et al. (2009) and Han (2014) also stressed that WVP and OP values decreased with increase of cellulose fibers in tapioca starch films. This behavior was associated with the stronger hydrophilic character of starch when compared with the cellulose fibers.

Table 2.7**: Water vapor permeability and oxygen permeability of potato starch composites (Hassani & Nafchi, 2014)**

Clay (%)	WVP × 10^{11} (g m^{-1} s^{-1} Pa^{-1})	OP (cm^3 μm^{-1} $(m^2\ day)^{-1}$)
0	2.91 ± 0.53	76.88 ± 2.44
1	2.14 ± 0.29	61.72 ± 1.28
2	1.44 ± 0.24	50.98 ± 1.02
3	1.02 ± 0.12	45.77 ± 0.50
5	0.83 ± 0.11	38.20 ± 0.81

CHAPTER 3

METHODOLOGY

3.1 Introduction

This chapter discusses about the main materials, equipment and method used for the preparation of starch/BP film. The properties of the films were investigated in term of morphology, mechanical properties (tensile strength (TS), Young's Modulus of elasticity (YM) and elongation at break (EAB)), optical properties (color analysis and transparency analysis) and barrier properties (water vapor permeability (WVP) and oxygen permeability (OP)).

3.2 Preparation of starch-based films incorporate with banana pseudo-stem powder

3.2.1 Raw material

All the material materials used in this work were food grade, edible and non-toxic. Tapioca starch (food grade) as in *Figure 3.1* obtained from LGC Scientific Sdn. Bhd. was the main ingredient used in the film preparation.

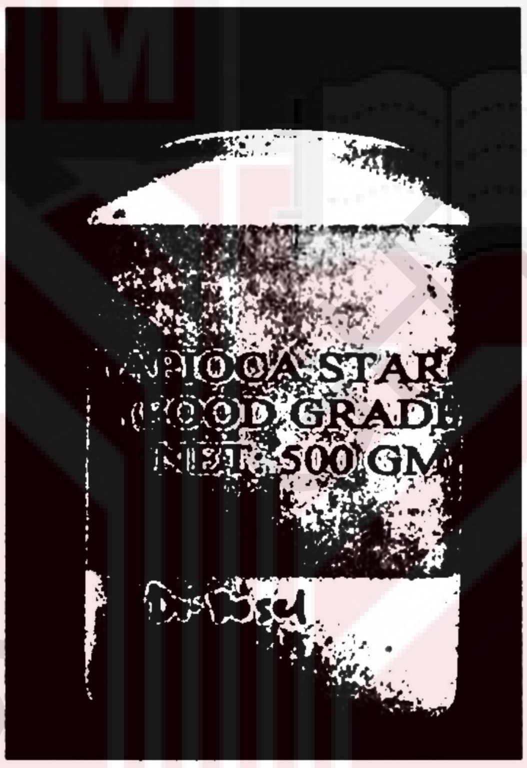


Figure 3.1: **Tapioca starch**

Banana pseudo-stem (*Musa acuminata X balbisiana ABB cv. Awak*) as in *Figure 3.2(a)* was obtained from local farm located at Lentang, Pahang. The stem was dried in the oven for 2 days and ground to form powder as in *Figure 3.2(b)*. The powder was kept in a desiccator (Temperature: 25°C, Relative Humidity: 53%) for further work.

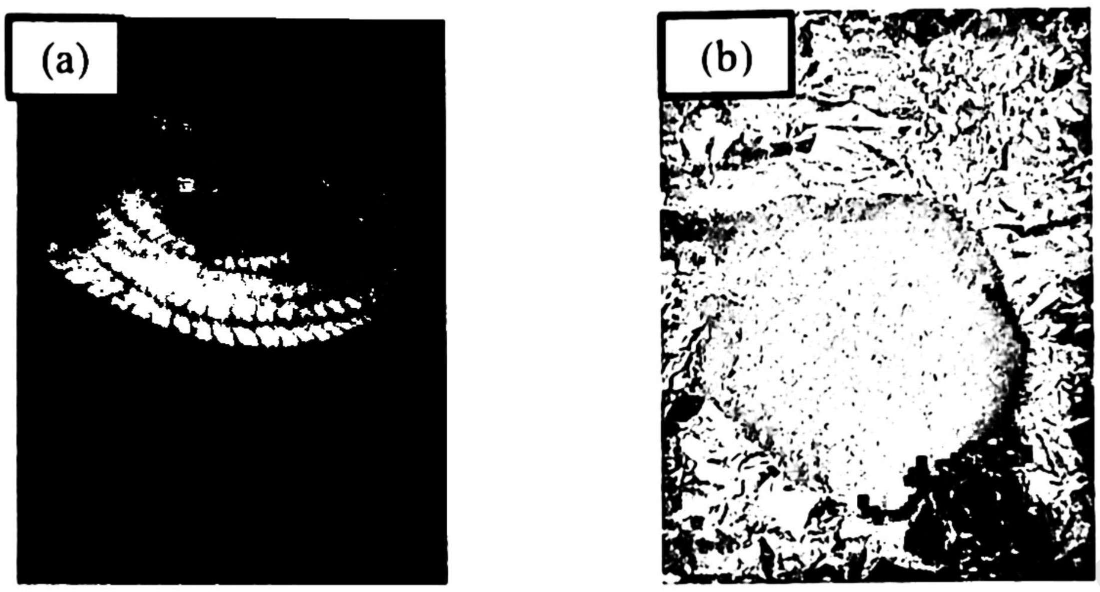


***Figure 3.2*: (a) Banana pseudo-stem and (b) banana pseudo-stem powder**

3.2.2 Banana pseudo-stem powder (BP)

The inner part of banana pseudo-stem was sliced manually using a sterile knife. The peeled pseudo-stem was rinsed with running tap water followed by rinsing with distilled water. The sliced of banana pseudo-stem was dried in a cabinet dryer at 60°C for 2 days and milled at 15,000 rpm with mesh pore size of 0.2 mm to obtain a fine BP. *Figure 3.3* shows the step of preparation of banana pseudo-stem powder.

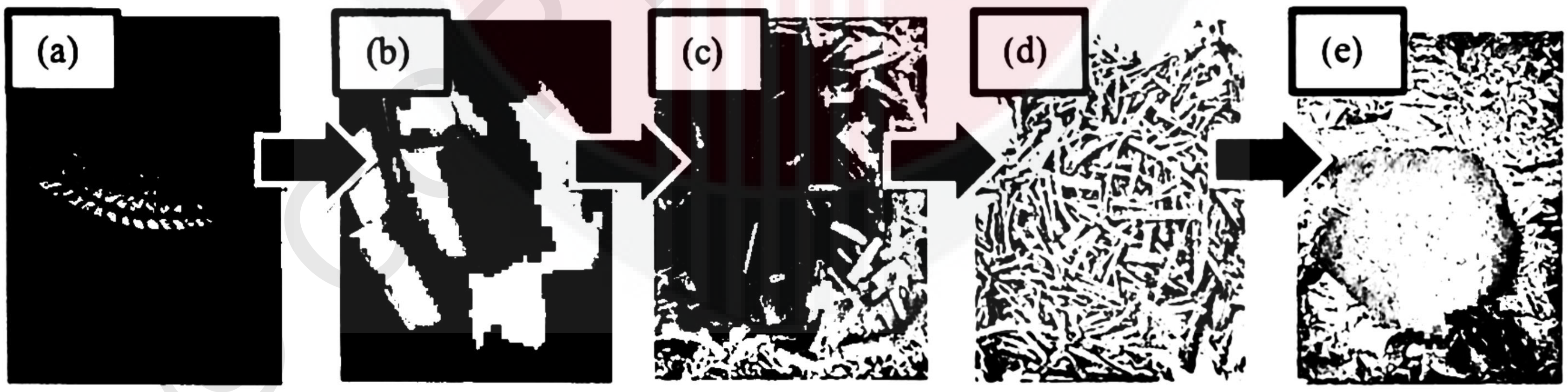


***Figure 3.3*: (a) Banana pseudo-stem, (b) peeled banana pseudo-stem, (c) sliced banana pseudo-stem, (d) dried banana pseudo-stem and (e) banana pseudo-stem powder**

3.2.3 Starch-based films incorporate with banana pseudo-stem powder

Thin films in the present work were prepared according to that of Shapi'i *et al.*, (2016) with some modification by different amount of BP instead of different concentration of chitosan. The amount of 3 g of tapioca starch was weighed using ER-120A Electronic Balance (Japan). The tapioca starch was added into 100 ml of distilled water to obtain a film forming solution. The mixture was stirred using Favorit stirrer hot plate (Model HS0707V2, Chicago) with continuous stirring for 5 minutes. Then, designated amount of BP was dispersed in the starch mixture. The percentage of the BP used was varied at 0, 10, 20, 30 and 40 wt. % on the dry basis of the tapioca starch. The mixture was stirred continuously and heated until the solution gelatinized at 70°C. An amount of 30 ml solution mixture was then spread evenly in a Petri dish (14 cm diameter) and dried at room temperature (28°C) for 48 hours. After dried, the films were peeled off and kept in a desiccator (Temperature: 25°C, Relative Humidity: 53%) prior to the characterization.

3.3 Characterization of the films

3.3.1 Morphological properties

The morphology of starch/BP films were observed using Scanning Electron Microscopy (SEM) (S-3400N, Hitachi, Japan) as shown in *Figure 3.4*. All the films were coated with gold before being observed under SEM. The test was carried out with voltage of 15kV.

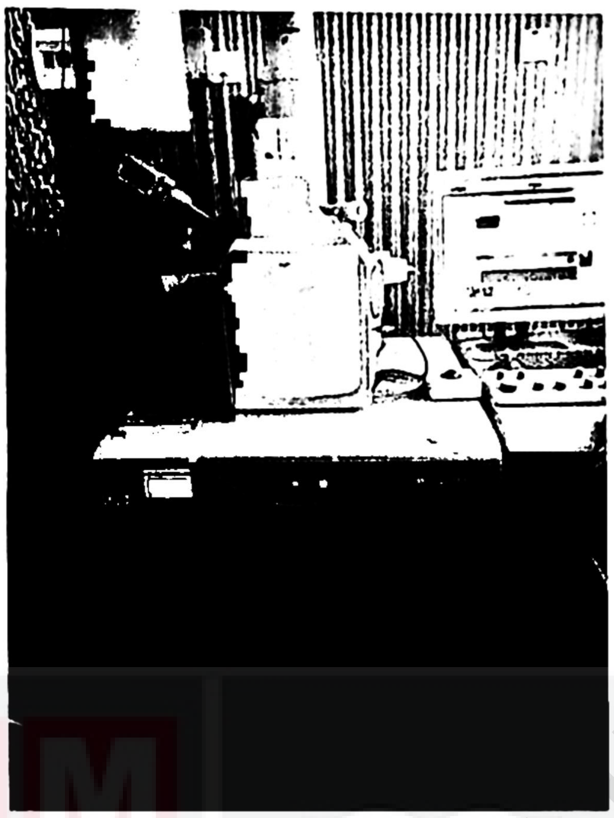


Figure 3.4: **Scanning Electron Microscopy**

3.3.2 Optical properties

The color and transparency of the films was determined using ASTM D1746 method (Sothornvit *et al.*, 2009). Both analyses were done using color spectrophotometer (HunterLAB, Ultra Pro, USA). The starch/BP film samples were cut into 100 mm×15 mm and placed directly in the spectrophotometer test cell. The test was carried out for at least three times.

Color analysis was determined using the International Commission on Illulination (CIELAB) color system with color parameters of L^*, α^* and b^* where:

L^* = (Lightness, 0= black), (100= white)

a^* = greenness (-), redness(+)

b^* = blueness (-), yellowness(+)

Transparency analysis is determined by measuring the light absorption at a wavelength of visible light between 400nm to 700nm using the spectrophotometer. *Figure 3.5* shows the spectrophotometer equipment used for optical analysis.

***Figure 3.5*: Color Spectrophotometer**

Total color difference (*ΔE**) of the starch/BP films (Mehran *et al.*, 2013) were calculated according to the following formula:

$$\textbf{Total color difference, } \Delta E = \sqrt{(\Delta L *)^2 + (\Delta \alpha *)^2 + (\Delta b *)^2}$$

where;

$L*$ = 0 (black) to 100 (white)

$\alpha*$ = –60 (green) to +60 (red)

$b*$ = –60 (blue) to +60 (yellow)

3.3.3 Mechanical properties

Texture analyzer (TA.XT2 Stable Micro Systems, UK) was used to investigate the mechanical properties of the starch/BP films as shown in *Figure 3.6(a)*. The film samples were cut into 100 mm × 15 mm using a film specimen cutter as shown in *Figure 3.6(b)* according to ASTM standard D882 (Qingjie *et al.*, 2014). The samples were stored in desiccators containing silica gel (Relative Humidity (RH): 53%) for one day at room temperature. The samples were then clamped between the grips of Texture Analyzer and the test was run with the clamp distance of 60 mm and a cross-head speed of 0.5 mm/sec respectively (Qingjie *et al.*, 2014).

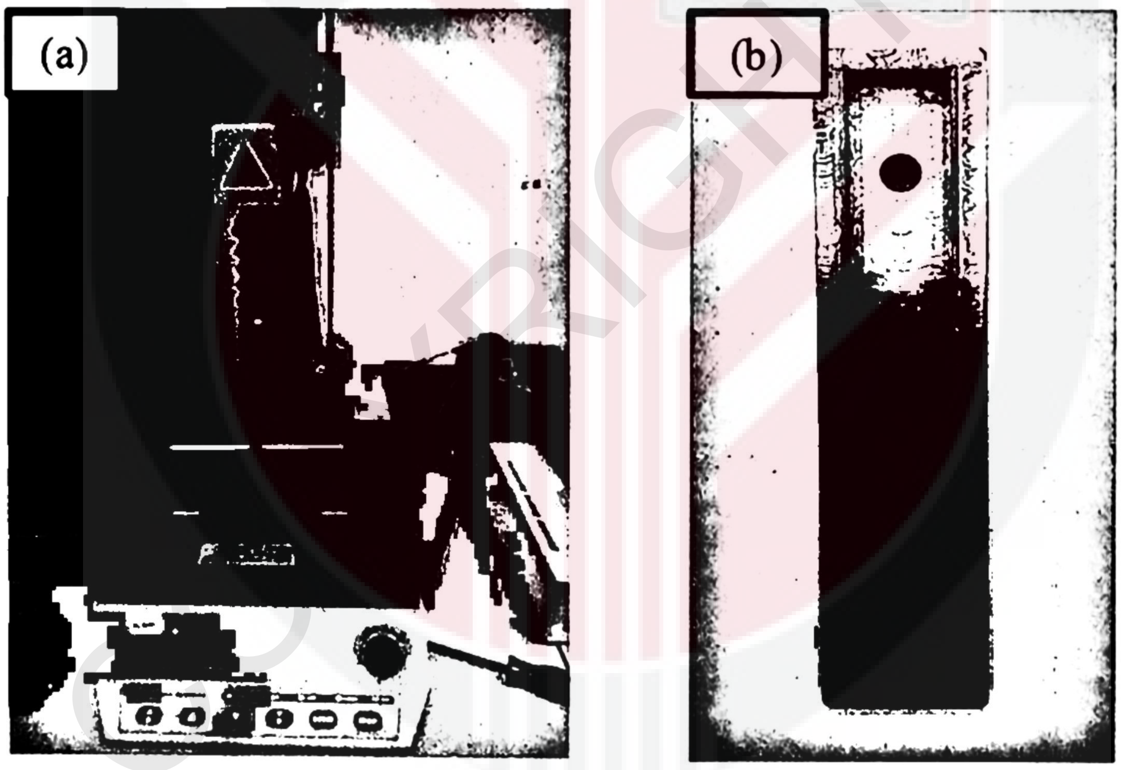


***Figure 3.6*: (a) Texture analyzer and (b) film cutter**

The mechanical properties include the tensile strength (maximum force used during measurement), Young's Modulus of elasticity (slope of the stress-strain curve at low values of strain) and elongation at break (ratio of elongation to original to the length of the sample) of the films. The general formula for tensile stress, σ (N/mm^2) is as the following (Hanpin and Stephen, 2013):

$$\textbf{Tensile stress, } \sigma = \frac{F_{max}}{A}$$

where;

F_{max} = maximum load (N)

A = cross sectional area of the sample (mm^2)

The tensile strain, ϵ of a film can be obtained using the following equation:

$$\textbf{Tensile strain, } \epsilon = \frac{\Delta l}{l}$$

where;

Δl = change in original length of sample

l = original length of sample

The general equation of Young's Modulus of elasticity, YM (MPa) is as the following (Hanpin and Stephen, 2013):

$$\textbf{Young's Modulus, YM} = \frac{\sigma}{\varepsilon}$$

The general equation of EAB is as the following (Hanpin and Stephen, 2013):

$$\textbf{Elongation at break, EAB} = \left(\frac{L_f - L_o}{L_o}\right) \times 100$$

where;

L_f = final length of sample

L_0 = original length of sample

3.3.4 Barrier properties

This analysis was done via gravimetric methods using water vapor permeability cup (Yasuda, No.318) as shown in *Figure 3.7*.

***Figure 3.7*: Water Vapor Permeability Analyzer**

The films were cut into circle shapes and sealed to the cup base with a ring using a combination of paraffin and bee wax solution (ratio 4:1). The thickness of the films was measured using Mitutoyo Digimatic Micrometer (Model MDC-1" SX). The amount of 5 g of anhydrous calcium chloride was put in the cup and placed in the desiccator at temperature of 30°C. Each cup was placed in a desiccator containing saturated magnesium nitrate to provide a constant RH 53%. The desiccators were placed in the incubator with the temperature control of 30°C. Weights of the cups were taken periodically every day until 10 days. WVP was calculated using the following formulas:

$$\mathbf{WVTR} = \left(\frac{w/t}{A}\right)$$

$$\mathbf{WVP} = \mathbf{WVTR} \times \frac{Thickness\ of\ film}{\Delta P}$$

where;

w/t = moisture gain weight per time (g/hr)
A = is the exposed surface area of the film (m^2)
ΔP = difference of partial pressures *(P_a)*

Oxygen permeation system equipped with a calorimetric sensor (MOCON, USA) was used to determine the oxygen permeability as shown in *Figure 3.8*. The test was carried out at room temperature of 27°C at a pressure of 3Psi according to ASTM F1927-07 (2007).

Before running the oxygen analysis, the thickness of the films was measured at five random positions and the average value was measured. Then, the oxygen transmission rate (OTR) of the films was determined for 50 cm^2 circular films using an optical measuring system. The film was put in the chamber and was clamped tightly.

***Figure 3.8*: Oxygen Permeability Analyzer**

Pure oxygen was introduced into the upper half of the sample chamber while nitrogen gas was injected into the lower half of the chamber. The measurement was repeated at least three times to ensure the accuracy of the result. Oxygen barrier was quantified by the oxygen permeability (OP), which indicates the amount of oxygen that permeates per unit of area and time. OP is correlated to the OTR by the following equation:

$$\mathbf{OP} = \frac{\boldsymbol{OTR}}{\boldsymbol{Thickness\ of\ film}}$$

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

This chapter presents the results and discussion on the findings of the present work, including the analysis and interpretation of data. Section 4.2 presents the results and discussion on morphological properties of banana pseudo-stem powder, starch film and starch/BP film. Section 4.3 presents the results and discussion on the optical properties of starch/BP films which include the color and transparency of the films. Section 4.4 presents the results and discussion on the mechanical properties of the films which include tensile strength (TS), elongation at break (EAB) and Young's Modulus (YM). Section 4.5 presents the barrier properties which were water vapor permeability (WVP) and oxygen permeability (OP) of starch/BP film.

4.2 Morphology properties

The scanning electron microscopy (SEM) was used to analyze the morphology of the banana pseudo-stem powder, control film and starch/BP film. *Figure 4.1(a)* shows the morphology of banana pseudo-stem powder. *Figure 4.1(b)* shows the morphology of tapioca starch film without the addition of BP while *Figure 4.1(c)* shows the morphology of 40% starch/BP film. (repetition)

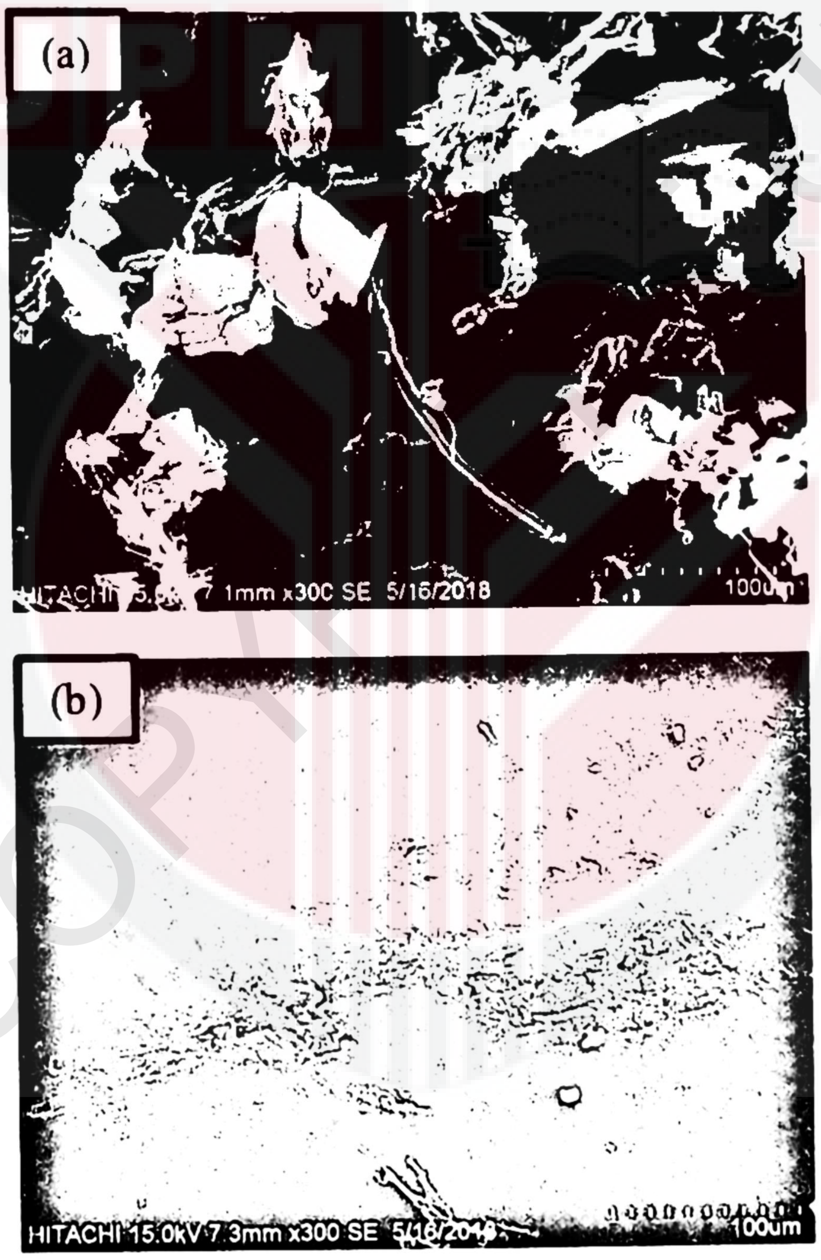


Figure 4.1: **SEM micrograph images magnification ×300 of (a) banana pseudo-stem powder, (b) neat starch film, (c) starch film incorporated with 40% BP**

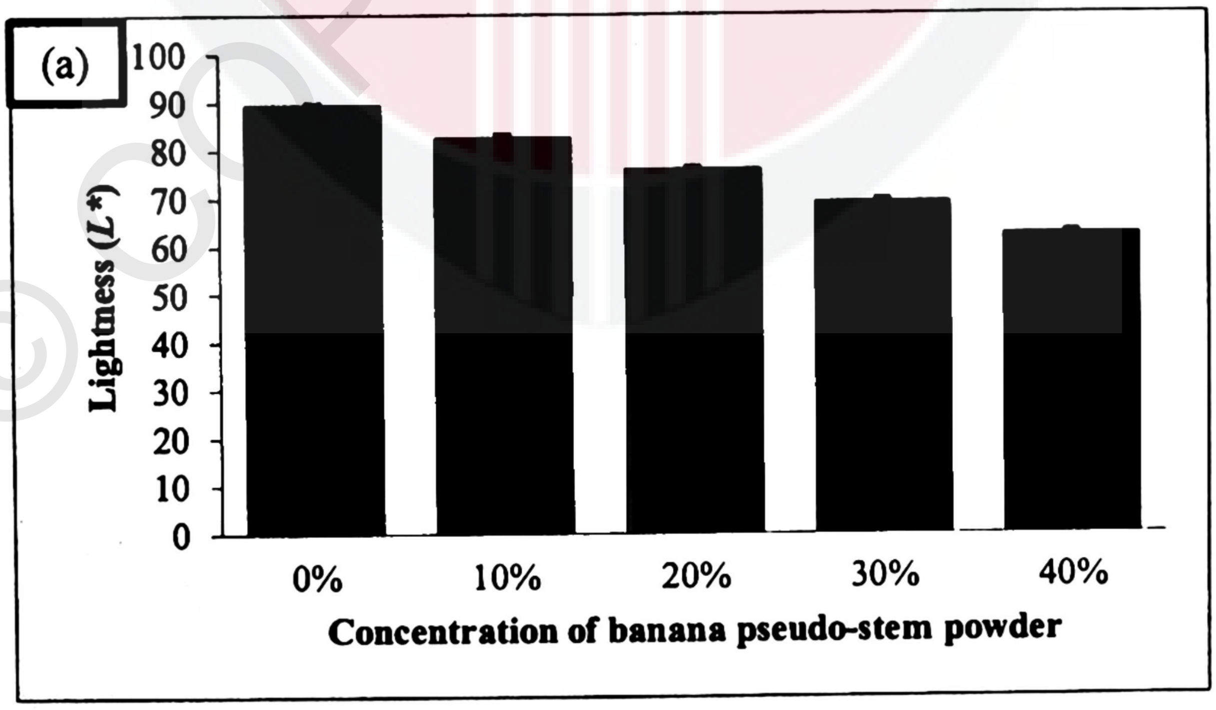
Figure 4.1(a) shows different shapes and sizes of BP powder when observed at 300× magnification. Flake-shape, fibrous and granular shape of different powder was observed by the SEM image. The particles were randomly positioned and the different shapes and sizes of the particles were due to the different components that existed in BP powder which include cellulose, hemicellulose, lignin, pectin, wax and extractives (Pappu et al., 2014). The SEM image of neat starch film (*Figure 4.1(b)*) shows a relatively smooth morphology of the film. This finding was supported by Wittaya (2009) whereby he also reported smooth surface morphology of neat starch film.

Meanwhile, comparing between SEM image of starch film incorporated with 40% BP (*Figure 4.1(c)*) and neat starch film (*Figure 4.1(b)*), it can be seen that 40% starch/BP film became rough with the addition of BP. This was because powder particles were distributed on the starch film surface. The difference in sizes and shapes of BP added into the starch film caused the poor dispersion and irregular arrangement of BP thus rough surface of neat starch film (Cheng *et al.*, 2017). The control film show smoother, homogeneous, uniform and compact surface with no pores. According to

Wang *et al.* (2017), the difference in the surface morphology of films were closely associated with the size and morphology of particles added into the film (Wang et al., 2017). The more similar size and regular morphology of the prepared particle size, the smoother and uniform the surface of the resulting films.

4.3 Optical properties

Color and transparency are the main properties that attributed customer attraction in food packaging application. The physical appearance of packaging film is important as it could affect the consumer acceptance for potential food packaging applications (Hosseini et al., 2013). Results of color and transparency analysis are presented below. Optical properties of starch/BP films were characterized in terms of lightness intensity (L^*), redness (a^*) and yellowness (b^*) as shown in *Figure 4.2(a)*, *(b)* and *(c)*.



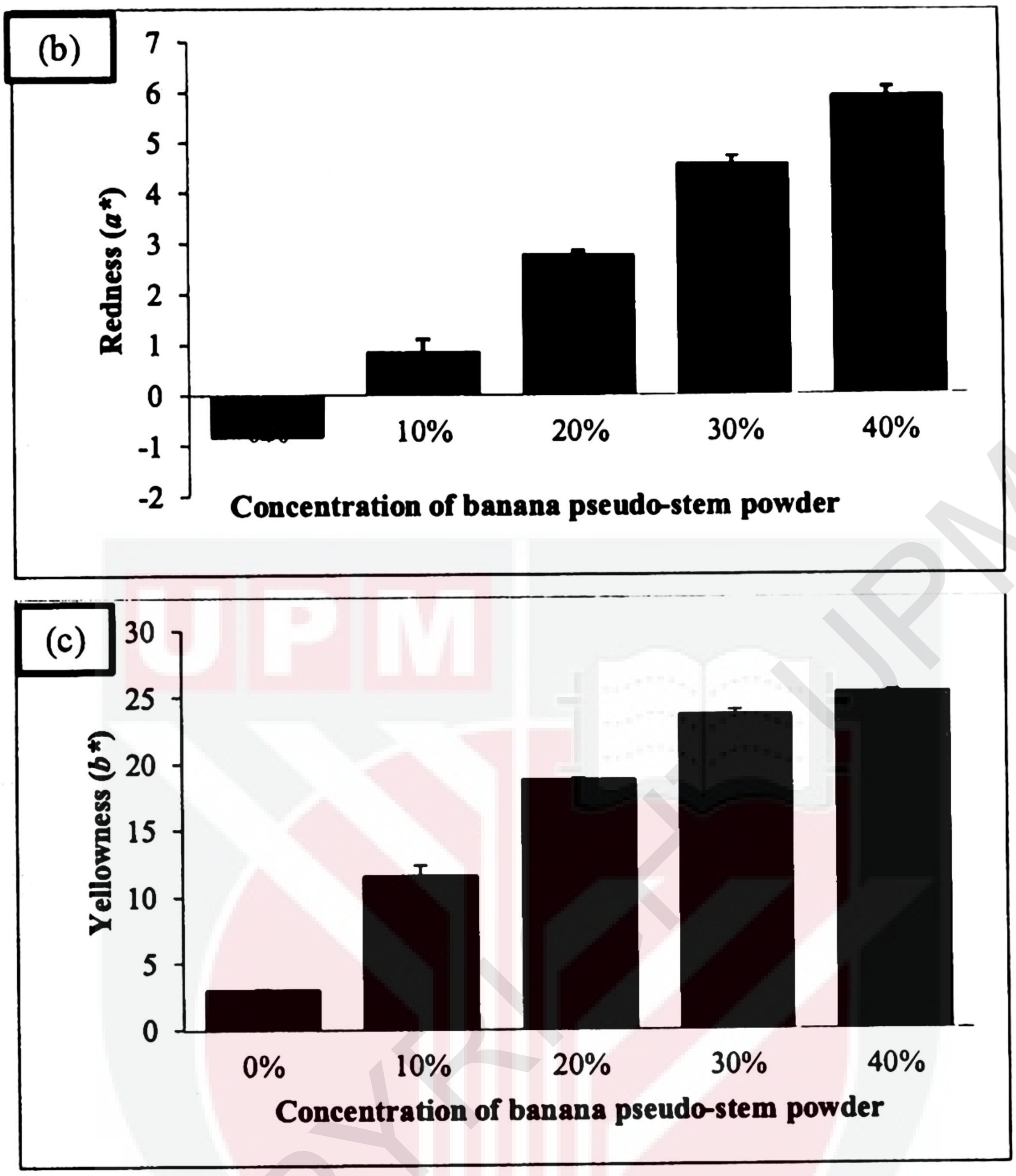


Figure 4.2: **(a) Lightness intensity (L*), (b) Redness (a*), and (c) Yellowness (b*)**

Figure 4.2(a) shows that starch films added with BP exhibited lower lightness (*L**) as compared to the control film which makes it good candidate for packaging film (Qingjie *et al.*, 2014). Lower lightness is a good packaging because it presentation encourages consumers to buy products. The decrement became prominent with the increase in concentration of BP. Film without the addition of BP exhibited the highest lightness because it was more colorless, translucent and clearer as compared to other film as observed during film preparation while film with 40% concentration of BP exhibited the lowest *L** value with percentage increment of about 30% compared to control starch film. The increase in the concentration of BP which was light brown in color resulted to the reduction in the lightness of the film.

Figure 4.2(b) shows an increment of redness (*a**) value for all starch/BP films. Control film shows negative *a** values with the value of –0.82 indicating that the films deviated towards green compared to other film due to the white nature color of tapioca starch. However, the *a** value of the control film was almost close to zero indicating that the green tone could be considered not noticeable. Meanwhile, the a* value increase with the addition of BP. There was a trend of increasing a* value with the increase in BP concentration indicating that the color of the starch/BP films were deviated towards redness resulted to the cloudy effects on the films. This was probably due to the color nature of BP which was light brown in color. The highest *a** value was film with 40% of banana pseudo-stem powder at *a** value of 5.93.

Figure 4.2(c) presents the yellowness (*b**) value of the films containing different concentration of BP. Control film exhibited the lowest *b** value compared to other films. The figure shows that the color of the films became yellower with the addition of BP. The increase in content of the BP in the starch film caused the increase in the b* value of the film. Again, addition of other biopolymer into the films particularly BP increase the cloudiness of the films (Hanpin *et al.*, 2013) which resulted to lower lightness and hence increased the *b** value of the film.

The total color changes (Δ*E**) from *Figure 4.3* also demonstrated that 10% BP/starch film exhibited the lowest Δ*E** value among other film with the value of 11.04%. Δ*E** values were observed to increase from 11.04 to 35.57% with respect to control film with the increase in concentration of BP from 10 to 40% consistent to the findings of L^*, a^* and b^* values. The differences might be related to the relative amount of the BP and the presence of small brown spot from BP which resulted to the cloudy effect (Qingjie et al., 2013).

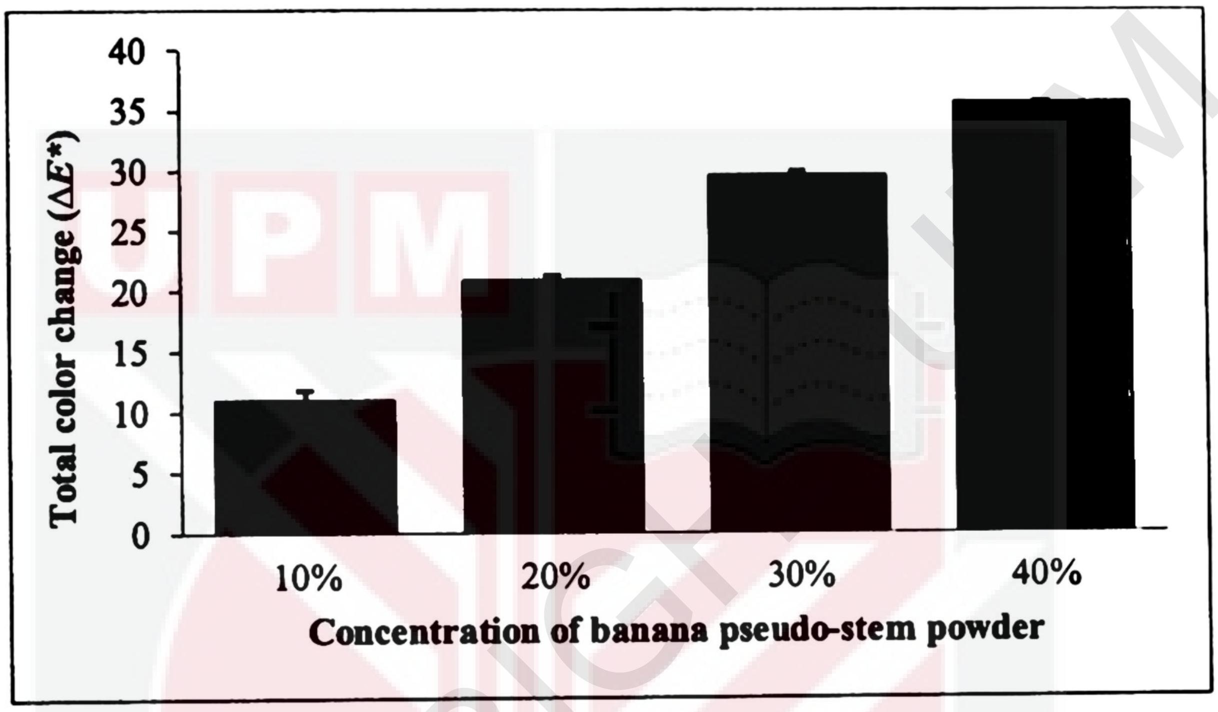


Figure 4.3: **Total color changes (ΔE^*)**

Transparency analysis was performed using light transmittance analysis in the spectra of visible region particularly at 600 nm. The results were shown in *Figure 4.4* for starch film added with different concentration of banana pseudo-stem powder. From the results obtained, control film proved to be the most transparent by showing the highest transmittance value of 75.9% in the spectra of visible region as tapioca starch is white color in nature. According to Lee et al. (2008), transparency of films can be affected by the homogeneity and amount of addition of other biopolymer. The addition of biopolymer would turn the control film into cloudier as a result of blending the pure film with other biopolymer powder (Riku, 2007).

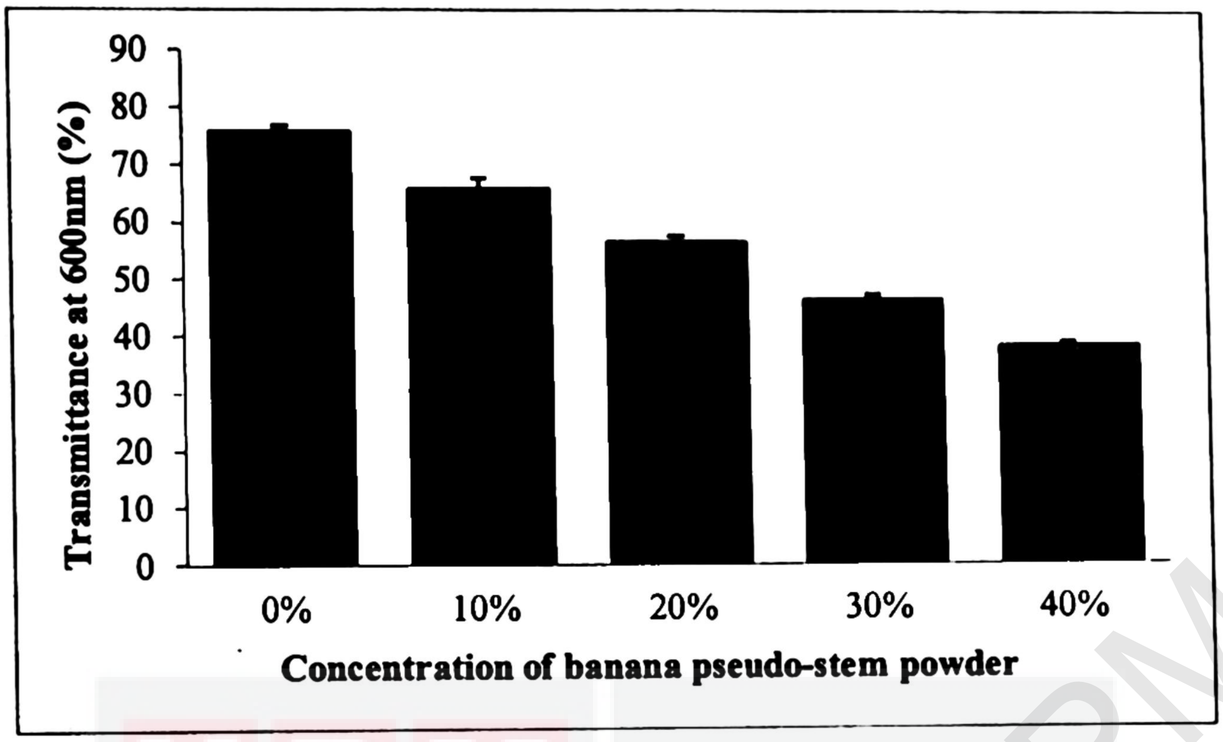


Figure 4.4: **Transmittance at 600 nm**

From *Figure 4.4*, it can be clearly seen that the transparency of the films decreased with the increase in amount of BP due to the higher solid particles content in the film which resulted in low transparency. This was because the large amount of BP in the starch matrix caused a blocking effect on the virtually transparent starch film in the visible spectra region (Lee *et al.*, 2008). The reduction in the transmittance might help to protect the packaged foods from visible and ultraviolet light that might lead to nutrient losses, discoloration and off-flavor (Arfat *et al.*, 2017). It should be noted that with the increase in BP powder concentration, the films became darker and thus exhibited lower transparency, which might also be attributed to the original color of BP added into the films.

4.4 Mechanical properties

TS plays an important role in determining the mechanical properties of films developed for use in many food applications because TS is an indication of the film strength (Javadian *et al.*, 2014). *Figure 4.5* shows the effect of different concentration of BP on TS of starch-based films.

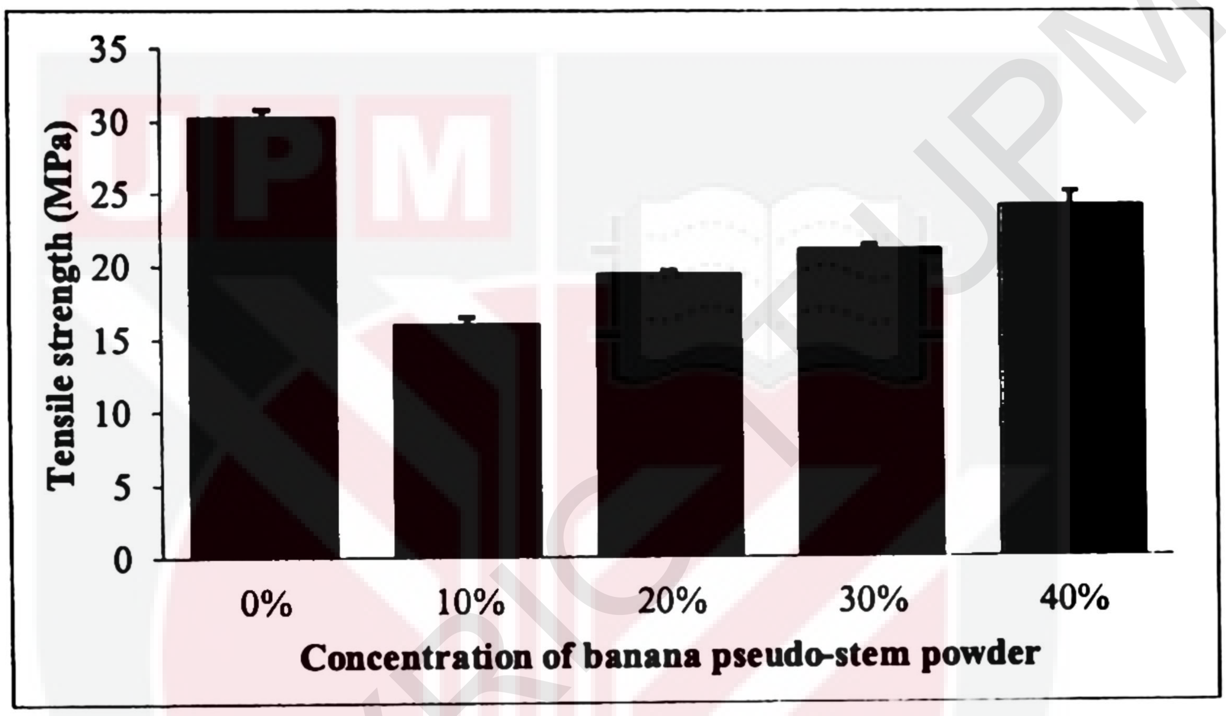


Figure 4.5: **Effect of different concentration of banana pseudo-stem powder on tensile strength of starch-based films**

The results obtained revealed that control film indicated the highest TS value which was 30.19 MPa. This was due to the strong intermolecular hydrogen bonds among the starch molecules (Wang *et al.*, 2015). The value of TS decreased with the addition of 10% BP by about 47%. This trend was presumably induced by the physical interaction between starch and BP. At 20% to 40% concentration of BP, TS value of films were slightly increased with the increase of BP concentration with the value of 19.47 MPa to 24.20 MPa due to the existence of cellulose in BP powder which acted as reinforcement in the starch matrix. However, the concentration of BP at 40% was not enough to produce optimum reinforcement whereby TS value of 40% starch/BP was still lower than control film.

Figure 4.6 shows the effect of different concentration of BP on YM of starch-based films. YM indicates the stiffness of the film. The low YM value indicate low stiffness of film which resulted to more flexible film.

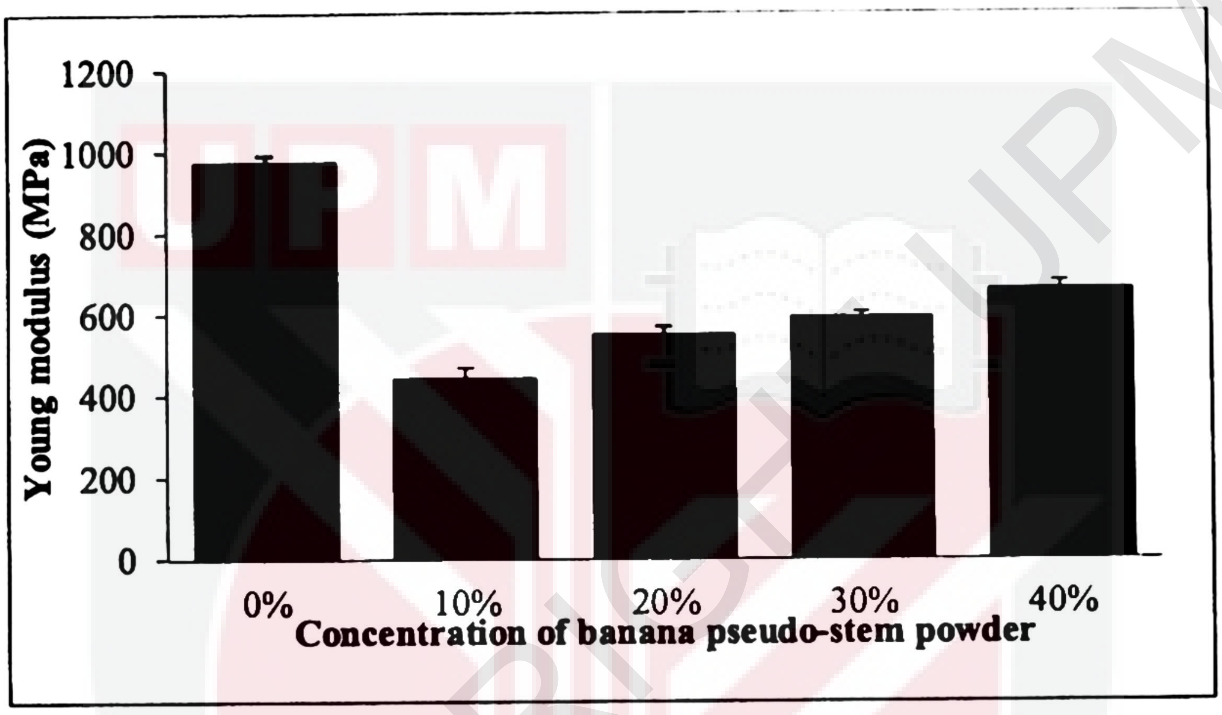


Figure 4.6: **Effect of different concentration of banana pseudo-stem powder on Young's Modulus of starch-based films**

Based on the *Figure 4.6*, the value of YM decreased with the addition of BP from 975.27 MPa to 393.02 MPa which was about 59% decrement as 40% of BP was added to the starch film. Low YM value indicated that the film exhibited high elasticity (Lee *et al.*, 2008). High value of YM at 0% concentration of BP indicated that the starch film exhibited brittle characteristic, easy to break and difficult to handle. The other concentration of BP showed a decrease in elasticity as compared to control film as can be seen from the decrease in YM value. This result demonstrated that films added with BP were less rigid and resistant, but were more flexible than the control film. The content of BP in starch matrix is important for film formation because the linear structure of amylose allows the molecules to orient parallel to each other, which favors the formation of hydrogen bonds gives rise to films with higher elasticity (Tako *et al.*, 2014). However, there was a trend of increasing YM with the increase in BP concentration which demonstrated that the elasticity reduced with the increase in BP content. This finding was consistent to the previous TS value findings. Increase in TS led to a decrease in elasticity of the films thus increase in YM.

Elasticity of the film was determined by quantifying parameter EAB (Lopez *et al.*, 2014). *Figure 4.7* shows the EAB values of starch films incorporated with different concentration of banana pseudo-stem powder.

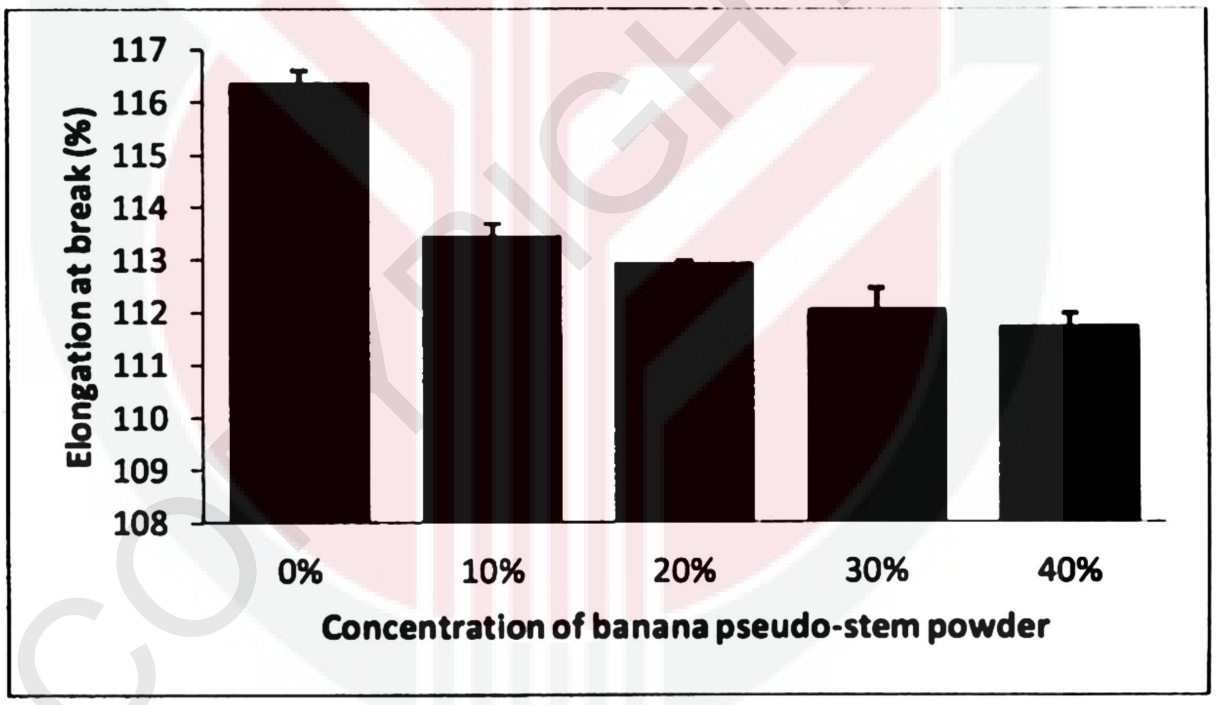


***Figure 4.7:* Effect of different concentration of banana pseudo-stem powder on elongation at break of starch-based films**

From *Figure 4.7,* it can be seen that the EAB value dcreased slightly with the addition of BP content in the film from 0% to 40% with the value of 116.35% to 111.71% respectively. According to Bangyekan (2006), the decrease in elongation was due to the interaction of starch-cellulose occurring at the molecular level which led to the strong adhesion between the biopolymer. The decrease in EAB value is an indication of the increase in the brittleness of the film (Voon *et al.*, 2010). Mechanical properties of the composite films have been reported to be highly dependent on the interfacial interaction between the biopolymer (Ma *et al.,* 2009). According to Voon et al. (2010), a decrease in EAB is desirable and advantageous for food packaging as it is directly related to the biodegradability of the film. When particle embedded in a network of starch polymeric chains in the matrix (Lu *et al.*, 2004) structural rigidity or brittleness of the bio-composite films are usually increased. Thus, the higher the brittleness of the films, the lower the EAB value.

Note that films added with BP concentration of higher than 40% was too brittle and did not formed well and thus mechanical properties of starch films added with BP concentration greater than 40% could not be determined. As a conclusion, 40% BP was the highest amount of BP that could be added to produce starch/BP film in order to minimize BP waste. 40% starch/BP film was the film that exhibited the best mechanical properties compared to other concentration of BP. In addition, the range of TS, YM, and EAB of 40% starch/BP film still fall within common commercial food packaging film type low density polyethylene (TS: 16.2MPa, YM: 300MPa and EAB: 68.7%) indicated that the films have the potential to be used as food packaging material.

4.5 Barrier properties

Water vapor and oxygen barrier properties are important for food packaging application to protect the food product from moisture and gas content that can lead to microorganism growth. These properties are important to be investigated to avoid or at least to decrease moisture and gas transfer between the food and the surrounding atmosphere. In this case, water vapor permeability and oxygen permeability of the produced films should be as low as possible.

The WVTR was calculated based on the slope of the linear regression line of the sample weight vs time graph whereby the slope was then divided by the area of the film being exposed to the transmission. The WVP was determined by multiplication of WVTR value with the film thickness and divided with the difference of partial pressure. The WVP of starch-based films incorporated with BP were examined at a vapor pressure difference across the films. The effect of BP powder addition on WVP of the starch-based films was shown in *Figure 4.8*. The WVP of starch-based films decreased with the addition of BP.

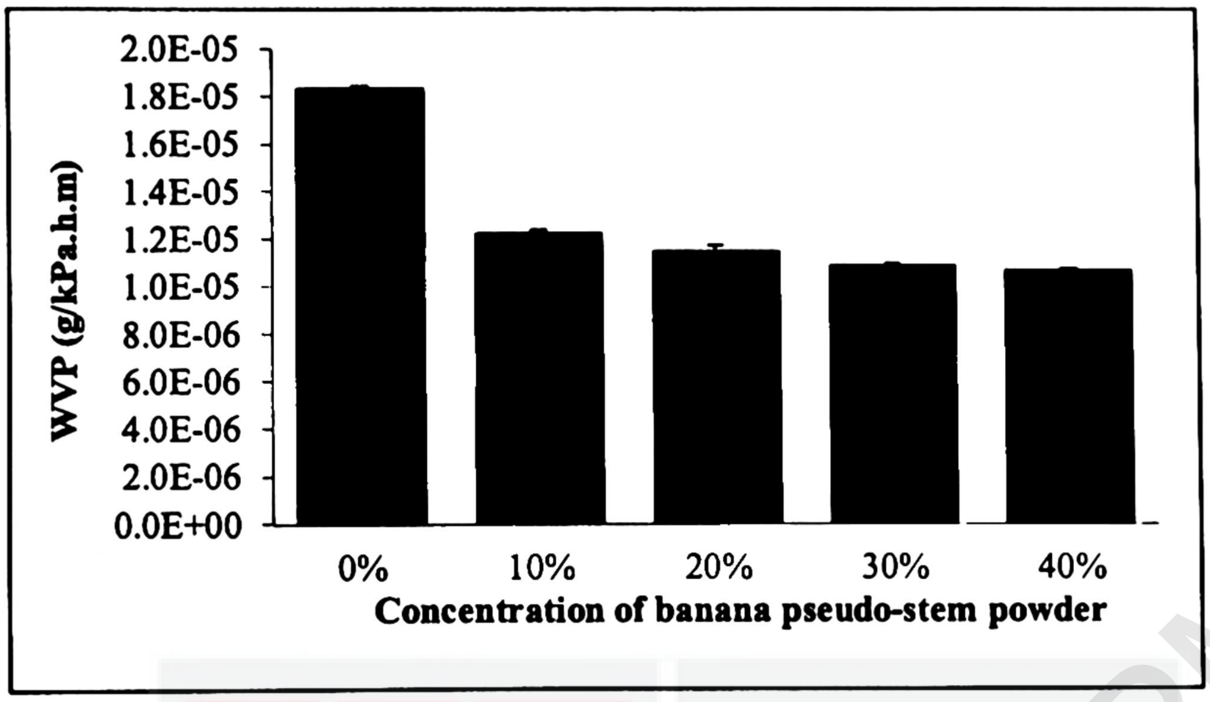


Figure 4.8: **Effect of different concentration of banana pseudo-stem powder on water vapor permeability of starch-based films**

The WVP value of control film was 1.83 × 10^{-5} g/kPa h m. The value decreased about by about 42% with the addition of 10% of BP into the strch film. However, the WVP of starch film decreased by about 44% with the addition of 40% BP. The WVP reduced because the diffusion of water vapor became slower with the addition of BP powder because the diffusion process was affected by the presence of impermeable particle which increase tortuosity in the film matrices (Sanchez-Garcia et al., 2008). Water molecules have to go through the tortuous path in the starch film thus lower the WVP values. The improvement of the water vapor barrier of the films was attributed to the formation of a rigid hydrogen bonded network of cellulose from BP in the composite that was governed by percolation mechanism (Voon *et al.*, 2010).

The amount of the oxygen gas that, over a given period of time at a steady rate, passes through a substrate was measured by oxygen transmission rate (OTR). The films for food packaging application must exhibit low oxygen barrier to avoid or reduce oxidative process during the transport, handling and storage of the food product. The oxygen permeability of the packaging film is important characteristic to be considered for a finished packing material to increase shelf life of foods (Miller and Krochta, 1997).

Figure 4.9 shows the starch films added with different concentration of against OP. The OP value of control film was 6.01 × 10^5 cc/m^2 day m. It can be seen that addition of BP in starch films reduced the OP of the starch film. Moreover, the higher the amount of BP added into the starch film, the lower the amount of oxygen that can pass through the film. The OP value decreased by about 27% with the addition of 40% of BP into the starch film. Addition of BP in tapioca starch films decreased the value of the oxygen permeability of the films due to high number of BP particles in the starch films whereby less oxygen was able to pass through the film at 40% of BP. The permeation of gas molecule was probably favored in the area between the starch and BP particles. Gas molecules were forced to diffuse around the starch in a random walk rather than taking a straight line path that lies perpendicular to the film surface due to the existence of impermeable BP.

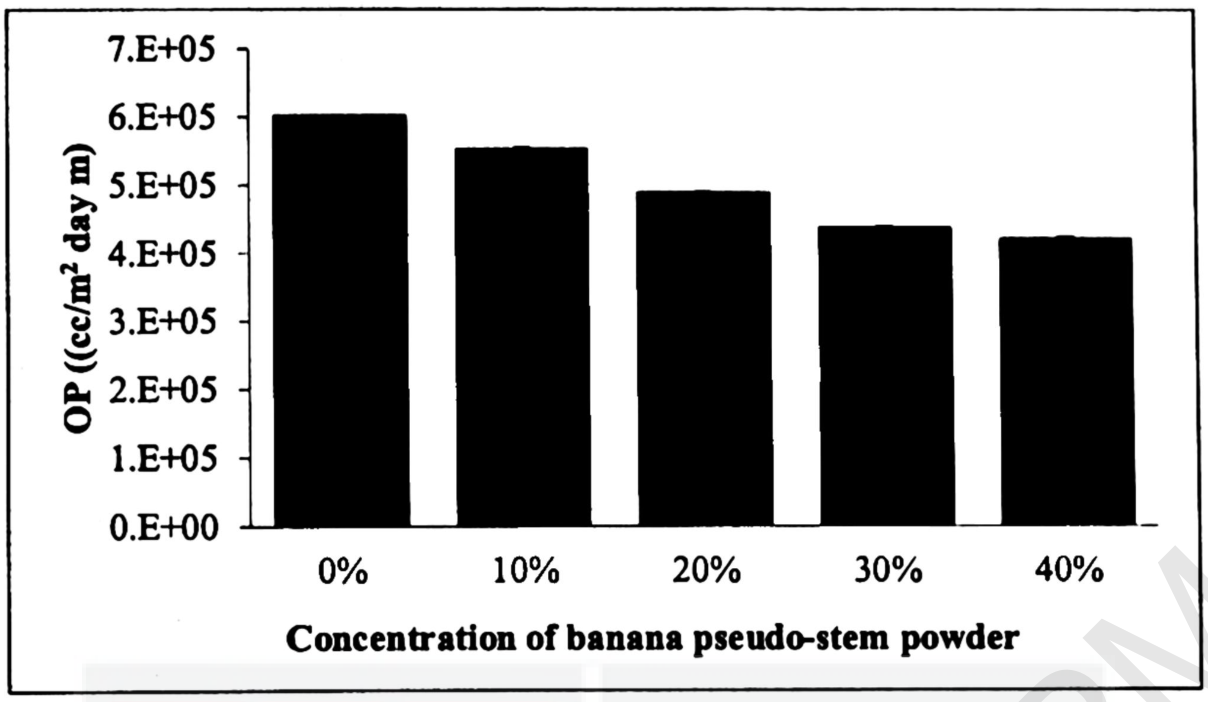


Figure 4.9: **Effect of different concentration of banana pseudo-stem powder on oxygen permeability of starch-based films**

The dispersion of BP in the starch film affected the permeability of the films. Starch has high oxygen permeability. The decrease in OP of starch films with the addition of BP was due to the generation of a more tortuous path for the permeation of oxygen molecules due to the presence of well dispersed BP in the starch biopolymer matrix (Voon *et al.*, 2010). Thus diffusion of the oxygen into the film decreased relative to the amount and orientation of the BP in the matrix. The state of aggregation and dispersion of BP in the film matrix are the importance factor that can affect the permeability of the film (Bharadwaj, 2001). Improvement in gas barrier properties was due to the concept of the tortuous path of the material.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

Tapioca starch films incorporated with different concentration (0, 10, 20, 30 and 40 wt. %) of BP were prepared by casting method. There were several analysis conducted on the films that were morphological, optical (color and transparency), mechanical (TS, YM and EAB) and barrier (WVTR and OP) analysis.

For color analysis, there was a slight decrease in L^* values but slight increase in a^* and b* values and ΔE values when BP was added into tapioca starch film. For transparency analysis, the values of the transmittance decreased with the increase in the amount of BP concentration in the starch film. This indicated that addition of BP to starch films led to changes in color and reduction in the transparency of the starch films. The starch/BP films changed in color and became less transparent with the increase in concentration of BP due to the light brown color in nature of BP powder and blocking effect by BP on the starch film respectively.

For mechanical analysis, tensile strength and the Young modulus increased with the increase in concentration of the BP up to 40 wt%. The best mechanical performance in tensile strength, elongation at break and young modulus was when the 40% BP was added into the starch film. Although the mechanical properties was lower than control films, the TS, YM, and EAB values (TS: 24.20 MPa, YM: 659.69 MPa, EAB: 111.71%) are still acceptable as compared to commercial packaging material which is low density polyethylene (LDPE) (TS: 16.2 MPa, YM: 300 MPa, EAB: 68.7%). Due to this, 40% concentration of BP was chosen as the maximum concentration of BP to produce starch/BP films in order to maximize the utilization of BP waste.

For barrier properties, it was found that WVP and OTR of starch films increased with the increase in concentration of BP powder due to the increasing of the BP content in the starch films. The diffusion of the water and oxygen became slower with the increase in BP powder due to the increase in tortuosity of the film matrixes.

In conclusion, the properties of the BP/starch films were very much dependent on the concentration of the BP added into the films. It was found that, 40% BP added into the film was the maximum concentration that produced the good performance properties of the films. Starch/BP films produced in this study have the potential to be used as food packaging material as their properties are still fall within the range of commercial packaging material.

5.2 Recommendations

It is recommended to utilize different biopolymer based other than starch such as PLA to produce the packaging film as well as using a different concentration of banana pseudo stem. Further investigation with addition of different types of plasticizers into the film is also recommended in order to produce efficient potential food packaging films. Besides, it is recommened to extract the cellulose from banana pseudo-stem before incorporated in film base in order to improve the properties of the films because this will ensure the consistency and purity of cellulose incorporated into the films. It is recommended that further study should be explored on other properties of the films such as thermal and antimicrobial properties. Another recommendation is to add other additives such as bleach to enhance the transparency of the starch/BP films. Finally, it is recommended to demonstrate the application of the film as potential food packaging material in future work.

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APPENDICES

APPENDIX A: DATA OF OPTICAL PROPERTIES OF FILM

Table A1: Lightness (*L**) of different concentration of banana pseudo-stem powder on starch-based film

Sample	Lightness, *L** (%)				
	1	2	3	Average	SD
0%	89.82	89.66	88.36	89.28	0.80
10%	83.76	82.98	80.88	82.54	1.49
20%	75.5	76.92	75.71	76.04	0.77
30%	69.91	68.99	67.72	68.87	1.09
40%	62.94	62.8	61.54	62.43	0.77

Table A2: Redness (*a**) of different concentration of banana pseudo-stem powder on starch-based film

Sample	Redness, *a** (%)				
	1	2	3	Average	SD
0%	-0.87	-0.83	-0.76	-0.82	0.05
10%	1.28	0.39	0.85	0.84	0.45
20%	2.71	2.72	2.9	2.78	0.11
30%	4.32	4.6	4.81	4.58	0.25
40%	5.74	5.79	6.27	5.93	0.29

Table A3: Yellowness (*b**) of different concentration of banana pseudo-stem powder on starch-based film

Sample	**Yellowness, *b** (%)**				
	1	**2**	**3**	**Average**	**SD**
0%	2.79	3.05	3.11	2.98	0.17
10%	11.78	10	12.8	11.53	1.42
20%	18.65	18.97	18.62	18.75	0.19
30%	22.97	24.03	23.98	23.66	0.59
40%	25.41	24.99	25.54	25.31	0.29

Table A4: Transmittance (%) of visible region at 600nm of different concentration of banana pseudo-stem powder on starch-based film

Sample	**Transmittance (%)**				
	1	**2**	**3**	**Average**	**SD**
0%	76.92	76.6	74.19	75.90	1.49
10%	68.7	65.98	62.98	65.89	2.86
20%	55.33	58.35	55.87	56.52	1.61
30%	47.86	46.03	44.97	46.29	1.46
40%	38.92	38.73	36.92	38.19	1.10

Table A5: Total color changes (ΔE^*) of different concentration of banana pseudo-stem powder on starch-based film

Sample	**Total color changes, ΔE^* (%)**				
	1	**2**	**3**	**Average**	**SD**
0%	11.05	9.72	12.35	11.04	1.32
10%	21.67	20.70	20.35	20.90	0.68
20%	28.82	29.95	29.88	29.55	0.63
30%	35.75	35.31	35.66	35.57	0.23
40%	11.05	9.72	12.35	11.04	1.32

APPENDIX B: DATA OF MECHANICAL PROPERTIES OF FILM

Table B1: Tensile strength of different concentration of banana pseudo-stem powder on starch-based film

Sample	Tensile Strength (MPa)				
	1	2	3	Average	SD
0%	29.09	30.44	31.05	30.19	1.00
10%	15.11	16.04	16.9	16.02	0.90
20%	19.77	19.09	19.55	19.47	0.35
30%	21.09	21.77	20.56	21.14	0.61
40%	25.61	24.54	22.45	24.20	1.61

Table B2: Young's Modulus of different concentration of banana pseudo-stem powder on starch-based film

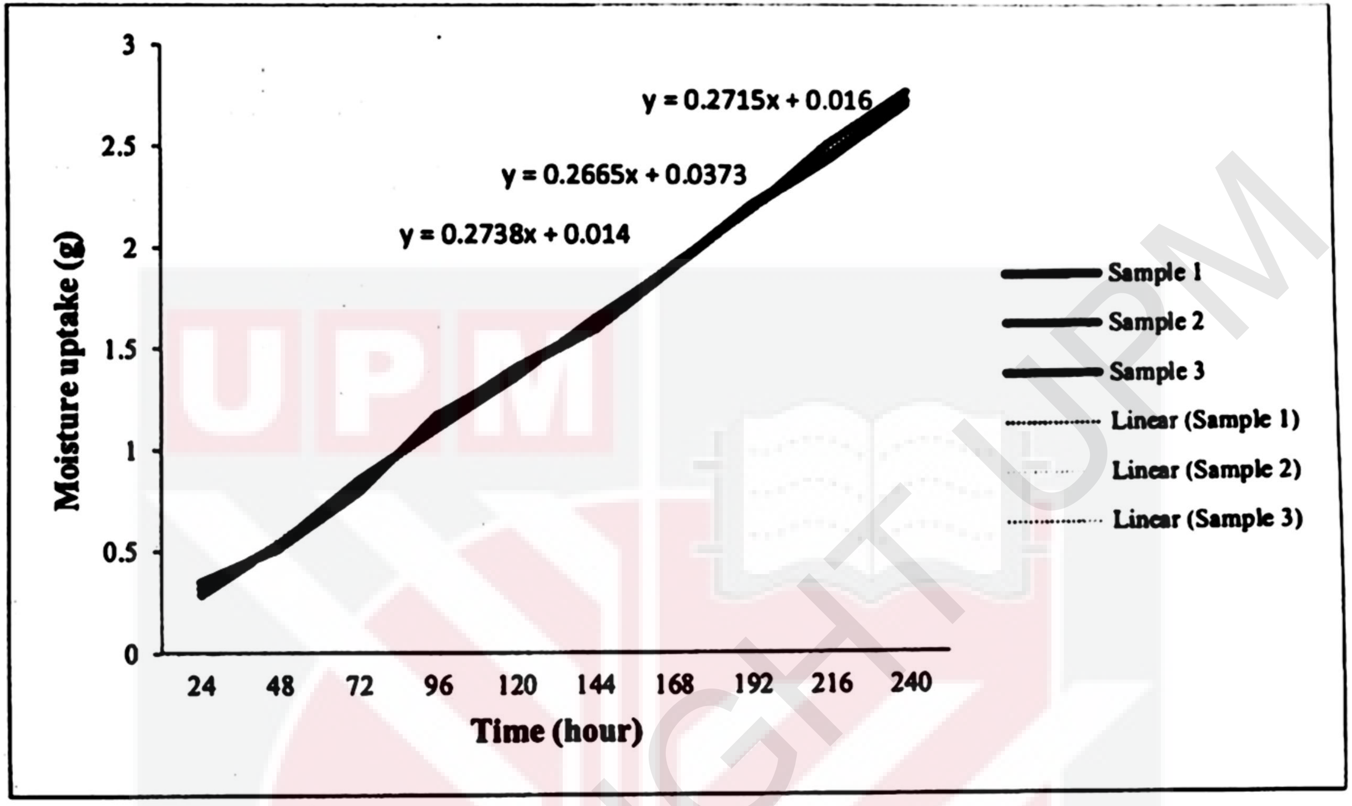
Sample	Young's Modulus (MPa)				
	1	2	3	Average	SD
0%	996.08	984.07	945.66	975.27	26.34
10%	400.55	491.05	435.77	442.46	45.62
20%	532.06	522.05	588.73	547.61	35.96
30%	569.55	605.06	599.60	591.40	19.12
40%	644.35	695.16	639.55	659.69	30.81

Table B3: Elongation at break of different concentration of banana pseudo-stem powder on starch-based film

Sample	Elongation at Break (%)				
	1	**2**	**3**	**Average**	**SD**
0%	116.02	116.21	116.82	116.35	0.418
10%	113.89	113.39	113.05	113.44	0.424
20%	112.95	112.99	112.86	112.93	0.064
30%	112.70	111.29	112.15	112.04	0.711
40%	111.53	111.39	112.21	111.71	0.442

APPENDIX C: DATA OF BARRIER PROPERTIES OF FILM

Graph C1: Moisture uptake of 0% concentration of banana pseudo-stem powder on starch-based film



Graph C2: Moisture uptake of 40% concentration of banana pseudo-stem powder on starch-based film

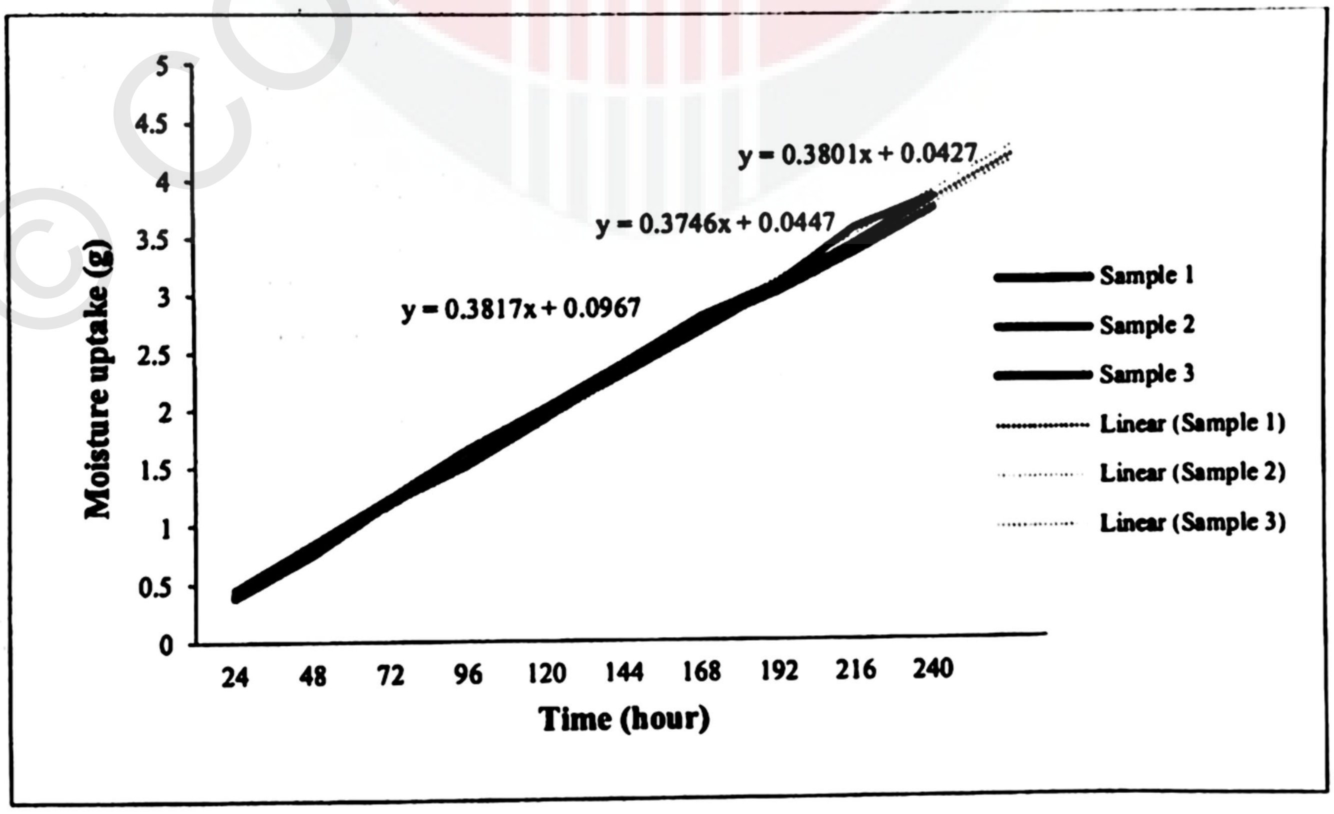


Table C1: Water vapor transmission rate and water vapor permeability of starch films at different concentration of banana pseudo-stem powder

Concentration of BP (%)	WVTR (g/m^2 h)	WVP (g /kPa h m)
0	96.64	1.83×10^{-5}
10	113.57	1.22×10^{-5}
20	123.09	1.17×10^{-5}
30	129.76	1.08×10^{-5}
40	135.39	1.06×10^{-5}

Table C2: Water vapor transmission rate and water vapor permeability of starch films at different concentration of banana pseudo-stem powder

Concentration of BP (%)	OTR (cc/m^2 day)	OP (cc/m^2 day m)
0	5.95×10^4	6.01×10^5
10	5.47×10^4	5.52×10^5
20	4.73×10^4	4.88×10^5
30	4.63×10^4	4.22×10^5
40	5.29×10^4	4.38×10^5