SYNTHESIS, CHARACTERISATION AND APPLICATION OF BIOPOLYMER FILMS INCORPORATED WITH NATURAL ADDITIVES FOR FOOD PRESERVATION

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SINTESIS, PENCIRIAN DAN PENGGUNAAN FILEM BIOPOLIMER DIGABUNGKAN DENGAN BAHAN TAMBAH SEMULA JADI UNTUK PENGAWETAN MAKANAN

ABSTRAK

Kajian ini menyelidik keupayaan filem biopolimer-bahan tambah untuk pengawetan makanan. Tiga biopolimer iaitu kitosan (CS), gelatin (GL) dan metilselulosa (MC) telah digunakan dalam kajian ini. Asid askorbik (AA), asid tanik (TA), minyak pati daun pisang (BA), minyak pati cengkih (CL), ekstrak kunyit (TU) dan ekstrak kamomil (CH) telah digunakan sebagai bahan tambah semula jadi. Kajian ini dibahagikan kepada lima kajian utama, iaitu sintesis, pencirian, aktiviti antimikrob, pengawetan makanan dan biodegradasi. Peralatan saintifik utama yang digunakan dalam kajian ini ialah spektrometer inframerah ransformasi Fourier (FTIR), mikroskop imbasan elektron (SEM), mesin ujian universal, penganalisis kebolehtelapan wap air (WVP), penganalisis kebolehtelapan oksigen (OP), spektrofotometer ultralembayung-nampak (UV-Vis) dan penganalisis termogravimetri (TGA). Bakteria yang digunakan untuk aktiviti antimikrob adalah *Staphylococcus aureus* (Gram-positif) dan *Escherichia coli* (Gram-negatif). Pengawetan sampel makanan telah dijalankan selama 7 dan 14 hari pada dua suhu persekitaran yang berbeza, iaitu 23-25 ˚C dan 27-30 ˚C. Tomato ceri (*Solanum lycopersicum var. cerasiforme*) dan anggur (*Vitis vinifera*) telah digunakan sebagai sampel makanan dalam kajian pengawetan. Dapatan kajian mendapati bahawa beberapa bahan tambah semula jadi telah berjaya menurunkan nilai WVP bagi GL-TA (1.73-1.28 g m^{-1} day^{-1} atm^{-1}), CS-TU (1.44-1.20 g m^{-1} day^{-1} atm^{-1}) dan MC-TA (1.27-1.18 g m^{-1} day^{-1} atm^{-1}). Dengan pengecualian penggabungan TA dengan GL, penambahan bahan tambah semula jadi mengurangkan kekuatan tegangan (TS) filem-filem biopolimer. Sementara itu, suatu kesan yang berbeza telah diperolehi bagi pemanjangan pada takat putus (EAB). Berdasarkan kajian aktiviti antimikrob, zon perencatan untuk CS terhadap *E. coli* telah meningkat daripada 10 hingga 25 mm dengan penambahan TU, manakala perencatan untuk CS terhadap *S. aureus* telah meningkat daripada 15 hingga 20 mm dengan rawatan BA. Semua filem-filem biopolimer yang telah digabungkan dengan bahan semula jadi telah dapat mengurangkan peratusan penurunan berat dan indeks pemerangan sampel buah-buahan. Kesimpulannya, penambahan bahan tambah semula jadi mengubah ciri-ciri fisikokimia filem CS, GL, dan MC yang membantu untuk memanjangkan jangka hayat makanan. Implikasinya, penggunaan filem biopolimer-bahan tambah semula jadi sebagai alternatif kepada filem berasaskan petroleum untuk pengawetan makanan boleh mewujudkan persekitaran hijau dan mampan.

ABSTRACT

This research investigated the potential of biopolymer-additive films for food preservation. Three biopolymers, namely chitosan (CS), gelatin (GL) and methylcellulose (MC) were used in this research. Ascorbic acid (AA), tannic acid (TA), banana leaf essential oil (BA), clove essential oil (CL), turmeric extract (TU) and chamomile extract (CH) were used as natural additive. This research is divided into five main studies, namely synthesis, characterisation, antimicrobial activity, food preservation and biodegradation. The main scientific instruments used in this study were Fourier transform infrared (FTIR) spectrometer, scanning electron microscope (SEM), universal testing machine, water vapour permeability (WVP) analyser, oxygen permeability (OP) analyser, ultraviolet-visible (UV-Vis) spectrophotometer and thermogravimetric analyser (TGA). The bacteria used for antimicrobial activity were *Staphylococcus aureus* (Gram-positive) and *Escherichia coli* (Gram-negative). The preservation of food samples was conducted for 7 and 14 days at two different surrounding temperatures, namely 23-25˚C and 27-30 ˚C. Cherry tomatoes (*Solanum lycopersicum var. cerasiforme*) and grapes (*Vitis vinifera*) were used as food samples in preservation studies. Research findings found that several natural additives have successfully decreased the WVP value of GL-TA (1.73-1.28 g m^{-1} day^{-1} atm^{-1}), CS-TU (1.44 -1.20 g m^{-1} day^{-1} atm^{-1}) and MC-TA (1.27-1.18 g m^{-1} day^{-1} atm^{-1}). With exception of incorporation of TA with GL, the addition of natural additives reduced the tensile strength (TS) of biopolymer films. Meanwhile, a contrast effect was obtained for elongation at break (EAB). Based on antimicrobial activity studies, the inhibition zone for CS against *E. coli* was increased from 10 to 25 mm following addition of TU, while the inhibition for CS against *S. aureus* was increased from 15 to 20 mm with BA treatment. All biopolymer films incorporated with natural additives were able to reduce the percentage of weight loss and browning index of fruit samples. In conclusion, the addition of natural additives changed the physicochemical characteristics of CS, GL, and MC films which favour to prolong the shelf-life of foods. In implication, the application of biopolymer-natural additive films as alternatives to petroleum-based films for food preservation could create a green and sustainable environment.

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LIST OF ABBREVIATIONS

AA	Ascorbic acid
BA	Banana leaf
BPA	Bisphenol A
CH	Chamomile
CL	Clove
CS	Chitosan
EAB	Elongation at break
FAO	Food Agricultural Organisation
FTIR	Fourier Transform Infrared
GL	Gelatin
LDPE	Low density polyethylene
MC	Methylcellulose
OP	Oxygen permeability
PVC	Polyvinyl chloride
SEM	Scanning electron microscope
TA	Tannic acid
TGA	Thermogravimetric Analyser
TS	Tensile strength
TU	Turmeric
WVP	Water vapour permeability

CHAPTER 1

INTRODUCTION

1.1 Research Background

The world population continues to grow, of which according to United Nations (2015) the world population has reached 7.3 billion as of mid-2015. Table 1.1 shows the projection of world population for the period of 2015-2100. As the world population is projected to increase, the market demand for food would continue to grow. As a human being, we need to consume food to stay alive and healthy. Since there is a great demand for food, food safety should be taken seriously. Food safety can be described as handling, preparation and storage of food in ways that prevent foodborne illness (Lim et al., 2016). Foodborne diseases have affected more than one-third of the total population in developing countries (FAO & WHO, 2002). In Malaysia, food poisoning cases tend to increase every year. For example, the rate of food poisoning in 2006 was 26.04 % and it doubled to 53.19% in 2007 (Lim et al., 2016). Furthermore, according to Hosseinnejad and Jafari (2016), Center for Disease Control and Prevention (CDC) of USA stated that 48 million people are infected, 128,000 are hospitalised, and 3000 are died in the United States each year due to foodborne disease.

Table 1.1

Projection of World Population for 2015, 2030, 2050 and 2100

	Population (millions)			
Major area	2015	2030	2050	2100
World	7 349	8 501	9 725	11 213
Africa	1 186	1 679	2 478	4 387
Asia	4 393	4 923	5 267	4 889
Europe	738	734	707	646
Latin America and the Caribbean	634	721	784	721
Northern America	358	396	433	500
Oceania	39	47	57	71

Note. Adapted from *United Nation*, 2015.

Food preservation is one of the oldest sciences used by human beings to keep food from polluted. In order to maintain the quality of the food and avoid food spoilage, preservatives are used to prevent or retard both chemical and biological deterioration of foods (Raj, Matche & Jagadish, 2011). The interesting fact about food preservation, it has been a part of every culture at nearly stage. Ancient people had to freeze seal meat on the ice in very cold climates and during tropical climates they dried foods under the sun. With food preservation, they had no longer to consume the kill or harvest immediately. Food preservation traditionally has three goals, namely: (1) the preservation of nutritional characteristics, (2) the preservation of appearance and (3) a prolongation of the time that the food can be stored (Abdulmumeen, Risikat, & Sururah, 2012). Traditional methods of preservation usually aim to exclude air, moisture, and microorganisms, or to provide environments in which organisms that might cause spoilage cannot survive (Abdulmumeen et al., 2012). As time passed, various methods are available for food preservation including drying, salting, canning and packaging (Wallace, 2005; Brody et al., 2008).

Nowadays, petroleum-based film or commonly called plastic is the material most used for food preservation. Plastic materials have been used in many applications since the late 1930s. Plastic packaging is the largest application for plastics about 40% in Europe, within the packaging niche, food packaging amounts to the largest plastics-demanding application (Fabra, Lagarón, Ocio, & Sánchez, 2016). Besides that, plastic materials as food and beverage packaging have gained increase importance during the last decade due to their ease of production, processing and low weight (Bott, Störmer & Franz, 2014). The advantages of plastic materials are chemically and mechanically resistant, lightweight, heat-sealable, can be printed on and are available in large quantities at low cost (Barbin et al., 2015). Although the use of petroleum-based materials is effective for food preservation, they are not degradable which can create serious environmental problems. Therefore, their use has been restricted to avoid further ecological damages (Tharanathan, 2003; Aider, 2010). Our earth greatly suffered from this pollutant which has caused environment destruction especially in marine habitat. Plastic pollutants are the major threats especially in marine environments where it assists in the transfer of the persistent organic pollutants (POPs) that may travel up the food chain (Corcoran et al., 2015).

Research in biopolymer have gained a lot of attention particularly in determining alternative ways to replace petroleum-based materials that potentially cause a negative environmental impact and problems associated to waste disposal (Dicastillo et al., 2016). Biopolymer is a material derived from plants or animals which is degradable and environment friendly. There are many types of biopolymer that have been used for food preservation such as starches, cellulose derivatives, chitosan/chitin, gums, protein and lipids (Elsabee & Abdou, 2013). Besides easily available, biopolymer-based materials can be used as layers to separate various food product, casing and coatings that are no need to remove for cooking and eating (Kowalczyk, 2016). It is interesting to note that petroleum-based materials are unable to be used as layers in food products due to they are not edible and toxic to human. However biopolymer materials as food preservatives have several disadvantages, where the main disadvantages is water sensitivity or poor barrier properties against water vapour that can be can be circumvented by combining them with lipids (Kowalczyk & Baraniak, 2014).

As discussed above, it is clear that there is an urgent need to find alternatives for petroleum-based films for food preservation. Both Department of Environment Malaysia and Ministry of Health Malaysia have highlighted this issue. Therefore, this research was devised as a direct response to the aforementioned issue.

1.2 Food Preservation Techniques

Food is any substance that can be eaten or drunk to serve nutritional support for the body or for pleasure (Abdulmumeen et al., 2012). It mostly contains of plant or animal origin which provide essential nutrients to consumers. Nutrients such as carbohydrates, fats, proteins, vitamins, or minerals are ingested and assimilated by an organism to produce energy, stimulate growth and maintain healthy life. Food preservation is the process of treating and handling food to stop or greatly slow down spoilage which leads to the loss of quality, edibility or nutritive value caused or accelerate by microorganisms (Abdulmumeen et al., 2012). However, some methods of food preservation use bacteria, yeasts or fungi to add specific qualities and to preserve food, for instance cheese and wine (Abdulmumeen et al., 2012). The preservative of food should be able to maintain or create nutritional value, texture and flavour which are important in preserving its values as healthy food. Moreover, preservation normally involves preventing the growth of bacteria, fungi, and other microorganism, and also retarding the oxidation of fats which cause rancidity.

Before the existence of commercial canning, freezing, refrigeration and freeze drying food, human being developed convenient methods to preserve fresh foods so that in the lean months they would have a relative constant food supply. In the beginning, drying, smoking, salting, fermentation, cold and potting were used. In later years, preservation using sugar, vinegar and alcohol were employed as preservatives. According to Raj et al., (2011), there are several traditional methods used to preserve food including thermal processing, drying, freezing, refrigeration, irradiation, modified atmosphere packaging and adding anti-microbial agents or salts.

Furthermore, several food additives such as calcium propionate, sodium nitrate, sodium nitrite, sulfites (sulfur dioxide, sodium bisulfite, potassium hydrogen sulfite, etc.) and disodium were used to preserve food (Abdulmumeen et al., 2012). According to Rahman (2007), the major food preservation techniques can be categorised based on the mode of action as: (1) slowing down or inhibiting chemical deterioration and microbial growth, (2) directly inactivating bacteria, yeasts, molds, or enzymes and (3) avoiding recontamination before and after processing. However, traditional preservation method may continue in one or more of these three ways. Figure 1.1 shows a number of methods from the aforementioned categories. In the following sections, several methods of food preservation are highlighted, namely drying, salting, canning and packaging.

1.2.1 Drying

Drying is regarded as a successful preservation method since it has been used across cultures and believed to be the oldest method (Harris & Taylor, 2004). Ancient people in the hot climates areas used this method to drying their food under the sun to preserve the food. That have proved where ancient Middle East and Oriental cultures actively dried their food in the baking sun to dry. The growth of microorganisms such as mold, yeast and bacteria occurred due to the high water content of food. Drying method can overcome issues related to microorganisms by reducing high volume of water in food, thus preserving them (Wallace, 2005; Harris & Taylor, 2004).

1.2.2 Salting

There is no single record that shows the exactly time when salt began to be used as a preservative method (Harris & Taylor, 2004). The ancient Egyptians, Chinese and early Romans preserve their meat by using salt and it is known that fish washed in seawater kept longer than fish washed in fresh water (Harris & Taylor, 2004). Fisherman in the Middles Ages kept their fish in barrels of brine or salt when their catch back to shore. Jams and jellies are preserved as solutions of high sugar content, and many meats (e.g., hams) and fish are still preserved by salting (Abdulmumeen et al., 2012). This method works due to the moisture in food is drawn out during the salting process which prevents bacteria to thrive.

1.2.3 Canning

During mid-eighteenth century, Napoleon's armies experienced difficulties to survive because of limited food which led to malnutrition and starvation. To solve the problem, Napoleon offered prizes to the first people who discover a reliable method for keeping France's armies fed. After fourteen years of experimentation, Nicolas Appert had discovered canning method for food preservation in 1795 (Abdulmumeen et al., 2012). He was the first who discovered successfully can meats, fruits and vegetables where he placed food in glass jars with wax sealed and reinforced with wire and then heating the jars. By using heat, microorganism responsible for the spoilage of food was destroyed and the expansion of the jar contents occur causes a greater pressure outside of the jar than the pressure inside which forming a vacuum. A seal is formed as the jar cools which preventing microorganisms from contaminating and entering the food.

Furthermore, Peter Durand, an English merchant, has modified Nicolas Appert's method to preserve food by using metal can. In fact, he commercialised the metal can as material for food packaging in 1910. However, during the earliest days of canning, a number of persons (including some Arctic explorers) have died as a result of exposure to the lead poisoning that was once used to solder cans (Abdulmumeen et al., 2012).

1.2.4 Packaging

Canning method that invented by Nicolas Appert was the first successful packaging (Harris & Taylor, 2004). In the beginning of the 20th century, tin-plated steel cans, glass bottle and wooden crates were used for food and beverage distribution (Brody et al., 2008). During World War II, wax and petroleum-based materials were used to protect ammunition became packaging materials for dry cereals and biscuits (Brody et al., 2008). Nowadays, the invention of packaging continues which led to the use of biopolymer-based materials and also active packaging in order to create a better food condition and replace petroleum-based materials.

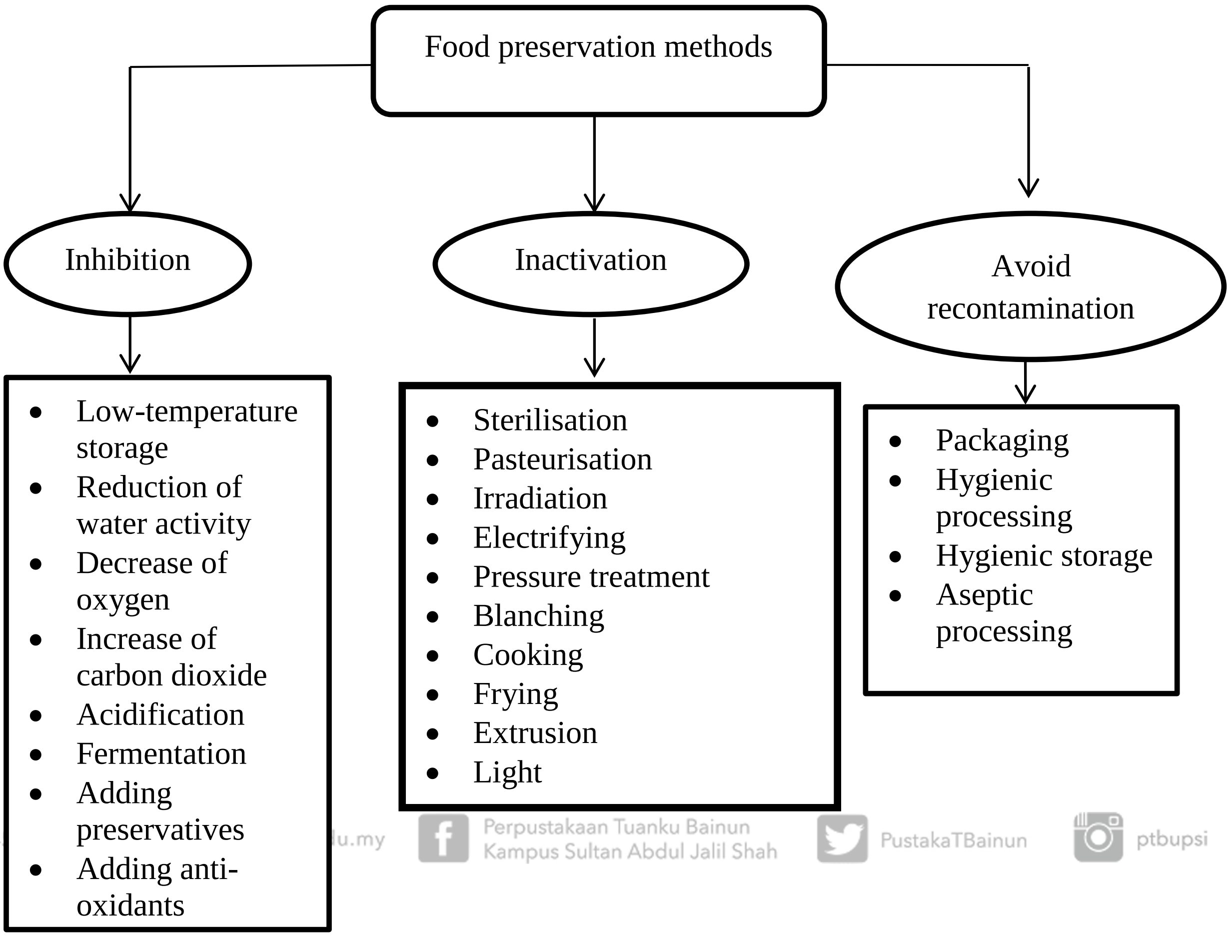

Figure 1.1. Major Food Preservation Techniques (Rahman, 2007)

1.3 Petroleum-material Packaging

Plastics or petroleum-based materials derived from crude oil are widely used as major materials for packaging that function as containing, protecting, preserving and transporting (Bilbao-Sainz et al., 2011; Science for Environment Policy, 2011). There are several petroleum-based materials used for packaging applications such as polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polyvinylchloride (PVC), polyamide (PA) and polystyrene (PS) because of their low cost, light weight, good mechanical and barrier properties, heat stability and easy to process (Hu, 2014). The global production of petroleum-based materials is estimated to be more than 200 million tons a year (Siracusa, 2016). Therefore, the problem regarding environmental issues rises due to the failure in waste management. Uncontrolled wastes of these materials polluting the marine environment and ingested by marine species or accumulated on the seabed, beaches and river estuaries (Science for Environment Policy, 2011). Furthermore, recycled of these polymers as packaging materials is the least compared to metals, papers, and glass, hence most of these materials will end up in the landfill sites and will remain for centuries before full degradation.

Besides, the recycled of packaging material wastes is uneconomical and impracticable due to contaminated with foodstuffs and biological substances (Hu, 2014). In addition to issues concerning the environment, the price of petroleum increased recently hence increases the price of petroleum-based material coincidently.

1.3.1 Types of Petroleum-material Packaging

The largest shares of plastic production are used as packaging materials in the Europe and at world level (European Commission, 2011). There are many types of plastics that have been used for packaging including low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP) and polyethylene terephthalate (PET). Petroleum-based materials have unique microstructure properties with many repeated unit chained together (Hu, 2014). These materials are able to be formed as bottle, bag, container, film, foam, coating, and wrapping in packaging industry. Table 1.2 shows the main polymers used in packaging applications.

Table 1.2

Polymers in Main Household Packaging Application

Application	Product	Polymer
Bottles	Dairy products	HDPE
	Juices, sauces	HDPE, PET, PP
	Water, soft drink	PET
	Non-food product (cleaning products, toiletries, lubricant, etc.)	HDPE, PET, PVC
	Medical products	PET
Closure	Caps and closures of bottles, jars, pots, cartons, etc.	PP, LDPE, HDPE, PVC
Bags and sacks	Carrier bags	LDPE, HDPE
	Garbage bags	HDPE, LDPE, LLDPE
	Other bags and sacks	LDPE, LLDPE, HDPE, PP
Films	Pouches (sauces, dried soups, cooked meals)	PP, PET
	Overwrapping (food trays and cartons)	OPP, OPS
	Wrapping, packets and sachets	PP, OPP
	Wrapping (meat, cheese)	PVDC
	Cling stretch rap film (food)	LLDPE, LDPE, PVC, PVDC

Note. Adapted from *European Commission*, 2011.
HDPE: high density polyethylene, PP: polyethylene, LDPE: low density polyethylene, PET: polyethylene terephthalate, PVC: polyvinyl chloride, LLDPE: linear low-density polyethylene, OPP: oriented polypropylene, OPS: oriented polystyrene, PVDC: polyvinylidene chloride.

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1.3.2 Impacts of Petroleum-material

According to Liu (2006), United States alone produced almost 230 million tonnes or 4.4 pounds per person of plastic wastes each day in 2001. Furthermore, Norway and Switzerland produced approximately 24.9 megatonnes of plastic waste in 2008 (European Commission, 2011). There were many effort that have been taken including reduce, recycle, reuse and recover to protect human health and the environment against harmful effects caused by the collection, transport, treatment, storage and landfilling of plastic wastes. Most of plastic packaging for food stuff is used for short-term but takes a long time to degrade. Therefore, plastics are easily found everywhere in society and also in the environment due to extensive production and the important uses in daily lives. However, the problems regarding plastic wastes start to risen up because plastic wastes are widespread with extensive accumulation in oceans, landfills and other terrestrial compartments which give bad effects toward wildlife and human health.

1.3.2.1 Environment

Nowadays, a large amount of plastic wastes has accumulated in the environment due the increase of production and the use of plastic. Plastic wastes had polluted the ocean, river and soil. These petroleum-based materials are not degradable which remain for many years. Science for Environment Policy (2011) reported that the layer of plastic wastes floating between the oceans of California and Hawaii has been estimated to span about 3.43 million km^2 (the size of Europe). Most of these pollutants may migrate and accumulate at shores. In 1992, a container ship lost 30,000 rubbers off the coast of China. Some of these turned up on the shores of the United Kingdom after fifteen years (Science for Environment Policy, 2011).

Numerous of wildlife becoming entangled in plastic waste which lead to injury or impaired movement and death (European Commission, 2011). According to Science for Environment Policy, (2011) a piece of plastic was found in an albatross stomach which cause from a crash of seaplane during World War II in 1944. Furthermore, Franeker et al., (2011) estimated that Northern Fulmars (a type of seabird) annually reshape and redistribute about six tonnes of plastic through ingestion of plastic waste every year. Besides, there is some evidence indicate that the toxic chemicals due to plastic ingestion which accumulate in living organism and throughout nutrient chains. Table 1.3 shows examples of known impacts on wildlife in terms of entanglement and ingestion.

Table 1.3

Number and Percentage of Marine Species with Documented Entanglement and Ingestion Records

Species group	Total number of species worldwide	Number and % of species with entanglement records	Number and % of species with ingestion records
Sea turtles	7	6 (86%)	6 (86%)
Seabirds	312	51 (16%)	111 (36%)
Penguins	16	6 (38%)	1 (6%)
Grebes	19	2 (10%)	0

(*Continue*)

Table 1.3 (*continued*)

Species group	Total number of species worldwide	Number and % of species with entanglement records	Number and % of species with ingestion records
Albatrosses, petrels, shearwaters	99	10 (10%)	62 (63%)
Pelicans, gannets, commorants, frigatebirds, tropicbirds	51	11 (22%)	8 (16%)
Shorinebirds, skuas, gulls, terns, auks	122	22 (18%)	40 (33%)
Other birds	-	5	0
Marine mammals	115	32 (28%)	26 (23%)
Baleen whales	10	6 (60%)	2 (20%)
Toothed whales	65	5 (8%)	21 (32%)
Fur seals and sea lions	14	11 (79%)	1 (7%)
True seals	19	8 (42%)	1 (5%)
Manatees and dugongs	4	1 (25%)	1 (25%)
Sea otter	1	1 (100%)	0
Fish	-	34	33
Crustaceans	-	8	0
Squid	-	0	1
Species total		136	177

Note. Adapted from *Science for Environment Policy*, 2011.

1.3.2.2 Health Risks

Plastic wastes can give bad impacts toward health of human and ecology because several plastics contain chemicals or additives to give certain properties. These chemicals are recently known to give bad effects on human and animal health which mainly affect the endocrine system. According to European Commission (2011), there are some proofs that indicate the toxicity of plastic from plastic ingestion which accumulate in living organism and throughout nutrient chains. There are several chemicals within plastics including Bisphenol A (BPA), phthalates and flame retardants (Science for Environment Policy, 2011).

Plastic industries used BPA as a harder and clear for polycarbonate food, beverage container and many other consume products (Galloway et al., 2010). It easily leaches at high temperature condition and causes a change in acidity (Ben-Jonathan, Hugo & Brandebourg, 2009). The consumption of BPA may results in chronic diseases such as cardiovascular and diabetes, as well as adult hormone dysfunction (Lang et al., 2008, Galloway et al., 2010). BPA can cause failure of reproductive system, increase in body weight and insulin resistance of animals (Ben-Jonathan et al., 2009). Oehlmann et al., (2009) stated that BPA is related to several diseases such as prostate cancer, breast cancer, sperm count decreases, miscarriage, obesity and diabetes.

Phthalate or diester of 1,2-benzedicarboxylic acid is an industrial chemical that normally used as a plasticiser for flexible or resilient which easily found in food packaging and other plastic materials (Meeker, Sathyanarayana & Swan, 2009).

Furthermore, it is easily to leach from plastic material to contaminate the environment because it is not chemically bound to the plastic matrix (Talsness et al., 2009; Meeker, Sathyanarayana & Swan, 2009). The exposure of phthalates causes negative effects on reproductive systems of human and animal, obesity and allergies (Meeker, Sathyanarayana & Swan, 2009).

Flame retardants such as polybrominated diphenyl esters (PBDEs) and tetrabromobisphenol A (TBBPA) cause disruption of hormone (oestrogen and thyroid) and damage of reproductive and nervous system (Science for Environment Policy, 2011). Besides, PVC and polystyrene have been found to release toxic monomers which linked to cancer and reproductive failure (Marcilla, Garia & Garcia-Queseda, 2004; Garrigos, Marin, Canto, & Sanchez 2004). These exposures may be due to water pollution and ingestion by animal which end up to the food chain. Therefore, plastic could cause most pressing problems such as cancers, climate change and food shortages in the future.

1.4 Problem Statement

The increase in production and use of non-biodegradable materials in food industry has polluted the environment. These types of materials are usually derived from petroleum products, which also caused waste disposal problem. Biopolymer materials have been used as alternatives to petroleum-based materials. However, biopolymer films have several drawbacks such as poor mechanical, thermal and barrier properties (Siti Hajar, 2014). Furthermore, biopolymer films possess a hydrophilic nature

(Shariatinia & Fazli, 2015). Although synthetic additives have proven improved the aforementioned properties, they are typically toxic and rise great concern among public consumers. In this context, natural additives have become ecologically important alternatives of synthetic additives (Wang, Marcone, Barbut, & Lim, 2012).

1.5 Research Gap

Based on literature review, although a number of studies had successfully developed and assessed the potential of biopolymer films for food preservation, several significant research gaps have been found and they are relevant to be investigated.

1. Pure biopolymer films (without additive) have several weaknesses such as low mechanical properties and poor water barrier properties. This limits their application as food packaging material. It is important to study the effects of incorporating additives on physical and chemical properties of biopolymer films.
2. Many of the previous studies reported the influence of only one type of additive to physical and chemical properties of biopolymer films. There is a need to compare the effects of several types of additives on physical and chemical characteristics of the films. In this study, three types of additives, namely pure compound, essential oil and plant extracts are used in the synthesis of biopolymer films.
3. Researchers normally used only one food pathogen in anti-microbial study. There is a need to compare the potential of biopolymer films to retard microbial growth from different type of bacteria or fungi. In this study, two

food-borne pathogens, namely *Escherichia coli* (gram-negative) and *Staphylococcus aureus* (gram-positive) are used to test the anti-microbial properties of biopolymer films.

4. The potential of biopolymer films to preserve real food products is not well studied. In this study, two local commodities were used namely cherry tomatoes and grapes as food samples. These fruits were wrapped by using biopolymer films in a preservation study for a period of fourteen days. The weight loss and browning index of the fruits were determined at the end of preservation study.
5. Up to date, the reports about biodegradation study of biopolymer films are scarce in public literature domains. It is imperative to conduct a biodegradation study of biopolymer films. In this study, the biodegradability of biopolymer films in soil environment up to six months period was evaluated.

1.6 Research Aim

The overall aim of this research is to develop environmental friendly films for food preservation. It is hoped that these films have great potential to be used as alternatives to non-biodegradable packaging films.

1.7 Research Objectives

The objectives of this research are:

1. To synthesis biopolymer-additive films using chitosan/gelatin/methylcellulose and natural additives.
2. To characterise the physical and chemical properties of chitosan/gelatin/methylcellulose films incorporated with natural additives.
3. To study the anti-microbial properties of chitosan/gelatin/methylcellulose films incorporated with natural additives
4. To evaluate the ability of chitosan/gelatin/methylcellulose films incorporated with natural additives to preserve fruits.

1.8 Research Significance

This research emphasises the significant of development of biopolymer films to replace or decrease the reliance of petroleum-based materials in food preservative. Moreover, this research highlights the efficacy of natural additives to provide active agents for the improvement of shelf-life and food safety of food.

1.9 Hypothesis

The addition of natural additives improves the ability of biopolymer films to preserve food products.

1.10 Thesis Organisation

Chapter 1 provides introduction of the study. In this chapter, several topics such as research background, history and method of food preservation, petroleum-based material, types of petroleum-based and impact of petroleum-based materials toward environment and health are outlined in order to better define the important of food preservation and the impacts of petroleum-based materials. Chapter 2 explains the biopolymer film, natural additives and the application of biopolymer film as food preservative. Chapter 3 describes a methodology in the synthesis of biopolymer films incorporating with natural additives and characterisation techniques in determine the chemical and physical properties of biopolymer films. Chapter 4 explains and discusses the findings of this research. The results include the chemical and physical characterisations, anti-microbial properties, ability to preserved food samples and biodegradation study of biopolymer films. Conclusions from this research and suggestions for future research are described in Chapter 5.