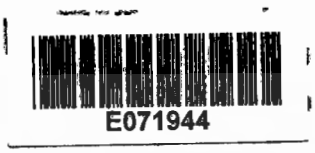
DEGRADABILITY OF FILM PREPARED FROM LDPE/MUCILAGE GEL OF OCIMUM CANUM SIMS



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Special Project Title: Degradability of Film Prepared from LDPE/Mucilage gel of *Ocimum canum Sims*

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Abstract

In this study, mucilage gel separated from *Ocimum canum Sims* seed was used as organic filler. Addition of plasticizer, glycerol or polyethylene glycol was studied. To blend LDPE with mucilage powder, two-roll mill was operated at 125°C for 15 minutes. Film of LDPE and mucilage was produced by blown film extrusion machine. The results were shown that tensile strength and modulus decreased as the percentage of elongation was increased when the mucilage powder and polyethylene glycol were increased in the blends. Water absorption of the blends were increased with an increment of mucilage powder content. The biodegradability of LDPE/mucilage blends have been studied in soil environment over a period of 4 months. Any changes in the various properties of the LDPE/mucilage before and after degradation were monitored using FTIR spectroscopy, an optical microscope (OM) for surface morphology, a thermogravimetric analyzer (TGA) for determine the decomposition temperature of mucilage powder content.

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Chapter 1

Introduction

1.1. Introduction

Nowadays synthetic materials like polyolefin are not degraded by microorganisms in the environment, which contribute to their long life and ever increasing volume of plastic waste. Some parts were reused, on the other hand some parts were disposal on the land or water resources, that is the problem for destroy it. To solve this problem we can controlled the usage of plastic product by use it not much more for necessity.

In recent times, biodegradable materials have gained importance particularly for the protection of the environment from ever-increasing plastic waste. Partially biodegradable polymers obtained by blending biodegradable and non-biodegradable commercial polymers can effectively reduce the volume of plastic waste by partial degradation. They are more useful than completely biodegradable ones due to the economic advantages and superior properties, imparted by the commercial polymer used as a blending component. One of the variable alternatives to accelerate the attack of microorganisms to LDPE is the addition of natural polymers, like starch, to guarantee at least a partial biodegradation. In these systems containing starch, the mechanical and rheological behavior, and also the susceptibility to degradation, will depend on various factors. For example, the amount and the nature of the starch used, which may be or not modified or gelatinized; type and concentration of additives, if used and processing conditions.

Starch and its derivatives are being a good choice since it is an abundant and low cost raw material in the market. Many studies in the literature have shown that this new material can be used for many purposes: domestic, industrial and agricultural, due to the facility on its processing.

Addition of a plasticizer such as glycerol can further improve the ductility of gelatinized starch (GS). Plasticized GS is known as thermoplastic starch (TPS) and is

capable of flow. Thus, mixtures of TPS with other polymers have the potential to behave in a manner similar to conventional polymer–polymer blend.

In this study, mucilage gel separated from *Ocimum canum Sims* seed was used as organic filler in LDPE film. For combination of LDPE and mucilage, glycerol or polyethylene glycol was added. Film of LDPE and mucilage were produced by blown film extrusion machine. The properties of film such as tensile, %elongation at break, and water absorption were investigated. TGA, MFI, OM and FT-IR were also used for characterization the blend film. To measure the decomposition, soil burial test was uesd.

1.2 Objectives

1.2.1 To study an optimum condition for preparation LDPE/mucilage blend film.

1.2.2 To design a new type of environmentally degradable plastic.

1.3 Scope of study

1.3.1 Prepare mucilage powder from *Ocimum canum Sims*.

1.3.2 Prepare film by blending LDPE with mucilage powder of *Ocimum canum Sims* by

- vary the weight ratio of mucilage powder and plasticizers.

- vary the concentration of mucilage powder, i.e., 1%, 5%, 10%, weight by weight of LDPE.

1.3.3 Characterizations of LDPE film blended with mucilage powder of *Ocimum canum Sims* by using TGA, FT-IR, MFI, OM, Universal testing machine, water absorption, and soil burial test.

1.4 Expected results

1.4.1 Can produce LDPE/mucilage blown film.

1.4.2 LDPE/mucilage film can decompose and degrade after soil burial test.

1.4.3 LDPE/mucilage film can used as packaging film and replace the typical film produced from pure LDPE.

Chapter 2

Theory and Literature Reviews

2.1. Low Density Polyethylene [1]

The first of the polyolefins, Low Density Polyethylene (LDPE) was originally prepared some fifty years ago by the high pressure polymerization of ethylene. Its comparatively low density arises from the presence of a small amount of branching in the chain (on about 2% of the carbon atoms). This gives a more open structure. Low Density Polyethylene (LDPE) is a most useful and widely used plastic especially in dispensing bottles or wash bottles. It is translucent to opaque, robust enough to be virtually unbreakable and at the same time quite flexible. Chemically LDPE is unreactive at room temperature although it is slowly attacked by strong oxidizing agents and some solvents will cause softening or swelling. It may be used at temperatures up to 95 °C for short periods and at 80 °C continuously. LDPE is ideally suited for a wide range of molded laboratory apparatus including wash bottles, pipette washing equipment, general purpose tubing, bags and small tanks. Dynalab Corp's plastic fabrication shop fabricates thousands of catalog and custom LDPE products.

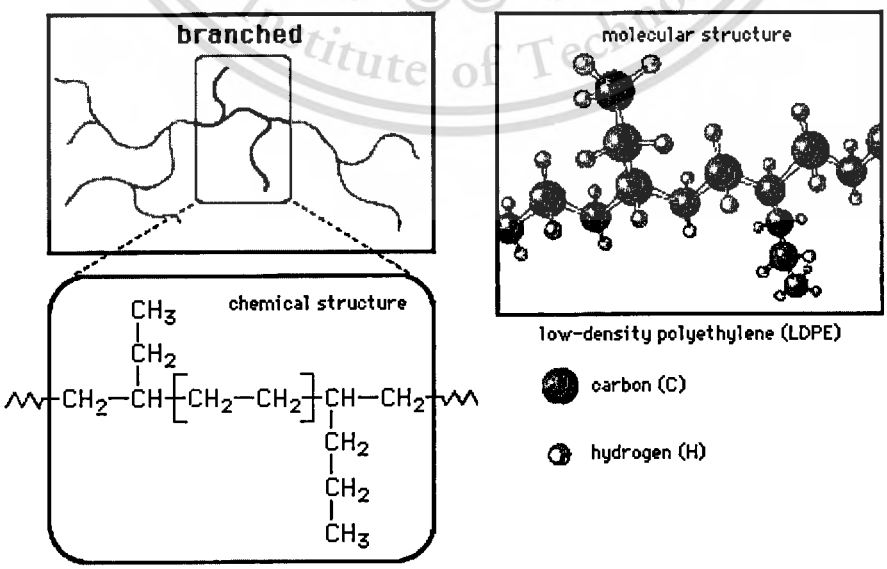


Figure2.1: Structure of LDPE [2]

LDPE is defined by a density range of 0.910 - 0.940 g/cm³. It is non-reactive at room temperatures, except by strong oxidizing agents, and some solvents cause swelling. It can withstand temperatures of 80 °C continuously and 95 °C for a short time. Made in translucent or opaque variations, it is quite flexible, and tough to the degree of being almost unbreakable.

According to more branching than HDPE, so its intermolecular forces (instantaneous-dipole induced-dipole attraction) are weaker, its tensile strength is lower, and its resilience is higher. Also, since its molecules are less tightly packed and less crystalline because of the side branches, its density is lower.LDPE is excellent resistance (no attack) to dilute and concentrated acids, alcohols, bases and esters and good resistance (minor attack) to aldehydes, ketones and vegetable oils. The limited resistances (moderate attack suitable for short term use only) of LDPE are aliphatic and aromatic hydrocarbons, mineral oils and oxidizing agents and poor resistance (not recommended for use with) is halogenated hydrocarbons.

Fabrication of LDPE include weldable and machinable, the parts that require flexibility, trays and containers, food storage and laboratory, corrosion resistant work surfaces, very soft and pliable etc. Applications of LDPE include chemically resistant fittings, bowls, lids, gaskets, toys, containers, packaging film and film liners, squeeze bottles, heat-seal films for metal laminates, pipe, cable covering, core in UHF cables, etc. Usage and properties of LDPE is similar to soft PVC.

2.1.1. Advantages and Disadvantages [3]

Advantages of LDPE include low cost, good chemical resistance, high impact strength at low temperatures, excellent electrical properties, can be processed by all conventional methods include plastic molding, plastic injection molding, extrusion, film blowing etc., and can be transparent in thin film form, etc.

Disadvantage of LDPE include the low strength, stiffness and maximum operating temperature, flammable, poor UV resistance, high gas permeability (particularly CO_2), susceptible to environmental stress cracking, etc.

2.2. *Ocimum canum Sims* [4]

An *Ocimum canum Sims* (botanical name) has english common name Camphor basil or Hairy basil. It is in Apiaceae (Labiatae) family.

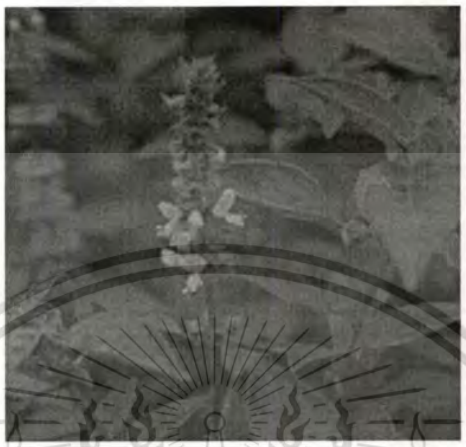


Figure2.2: *Ocimum canum Sims* [4]

An *Ocimum canum Sims* is bushy which has semi-woody herb to 40 cm high. It widespread in tropical Africa and old world tropics then introduced into the Americas.

The leaves are used as a traditional medicine in West Africa for the treatment of fevers, dysentery and to relieve toothache. Leaves used as flavoring and as an insect repellent. One percent (w/w) admixed with pinto beans caused 100 percent mortality in adult *Z. subfasciatus* after 48 hours. It is also used in Rwanda to protect against post-harvest insect damage.

Linalool is an oxygenated monoterpenoid which acts as a reversible competitive inhibitor of acetylcholinesterase. It is the major component of the oil, forming 60-90 percent of the total volatiles collected. Composition of the essential oil varies according to its origin. In East Africa it contains 16-25 percent camphor, while in Central Africa methyl cinnamate predominates; in West Africa it may contain 75 percent methylchavicol (estragol).

Table 2.1: Chemical components from each part of *Ocimum canum Sims* [5]

Leaves	Borneol, Camphene, Caryophyllene, p-cymene, Humulene, Myrcene, Nevadensin, Oleanolic acid, α-Pinene, Sabinene, trans- Sabinene hydrate, α-Selinene, Terpinolene, Ursolic acid, Methyl chavicol, Triterpenoid, Isoborneol, (+)-Camphor, 1,8-Cineol, β-Elemene, Limonene, Myrtenol, Oct-l-en-3-ol, α-Phellandrene, β-Pinene, cis-Sabinene hydrate, Salvigenin, β-Selinene, γ-Terpinene, Eugenol, Linalool, and Essestail Oil
Flower	Navadensin, Salvigenin, Oleanolic acid, and Ursolic acid
Seed	D-Arabinose, D-Glucose, D-Mannose, D-Xylose, D-, alacturonic , cid, Pentosans, Polysaccharide, Camphor, Oil, L-Arabinose, D-Galactose, L-Rhamnose, Galacturonic acid, D-Mannuronic acid, Mucilages, Camphene, and Myrcene
Scented oils	Bornel, α –Cadinene, β-Cadinol, Caryophyllene oxide, Methyl ether chavicol, Cinnamic acid methyl ester, β-Elemene, Estragole, Eugenol methyl ether, Geraiol acetate, Linelool, Myrcene, α-Pinene, Borneol acetate, β-Cadinene, Camphor, β-Caryophyllene, 1,8-Cineol, P-Cymene, Elemol, Geraniol, Limonene, Linelool acetate, Ocimene, abd Tricyclene

2.3. Mucilage [6]

Mucilage is slimy liquid cloudy-white jelly which separated from *Ocimum canum Sims* seeds. The easy steps to separated mucilage were shown in part of the experiment. The characteristic of mucilage is high viscosity even though in low concentration. When dry the mucilage, it change to thin white-brown film and this thin film can immediately swollen when contact to water.

The chemical component of mucilage is polyuronide. Polyuronide is a linear co-polymer of β-D-Mannuronic acid (M) and α-L-Guluronic acid (G), as shown in Figure 2.3. In nature, polyuronide is in form of salt, may be salt of calcium, potassium or manganese. Figure 2.4 and 2.5 showed the structure of sodium polymannuronic acid and sodium polyguluronic acid.

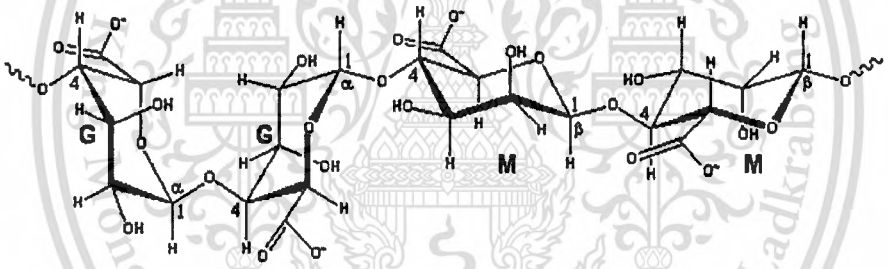


Figure 2.3 Polyuronide structure [6]

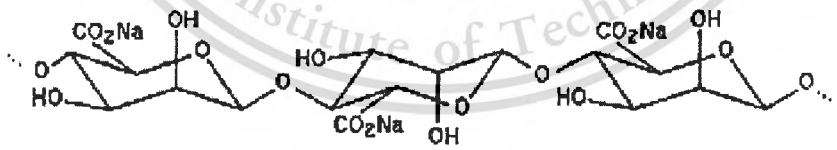


Figure 2.4 Sodium polymannuronic acid [6]

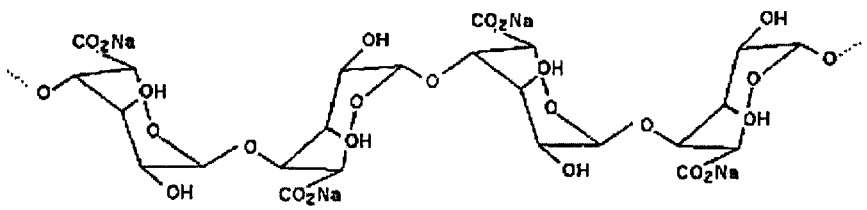


Figure 2.5 Sodium polyguluronic acid [6]

However, the mucilage from *Ocimum canum Sims* include sugar and uronic acid, connect by glycoside linkage. Sugar in structure may be pentose or heptoses or may be both of its. Uronic acid in structure that mostly found is glucuronic acid and galacturonic acid. Polygalacturonic acid, pectin, is a cementing material in the cell walls of all plant tissues. It is the methylated ester of, which consists of chains of 300 to 1000 galacturonic acid units joined with 1α→4 linkages. The degree of esterification (DE) affects the gelling properties of polygalacturonic acid. The structure shown in Figure 2.6 has three methyl ester forms (**-COOCH$_3$**) for every two carboxyl groups (**-COOH**), hence it is has a 60% degree of esterification, normally called a DE-60 pectin. Pectin is an important ingredient of fruit preserves, jellies, and jams.

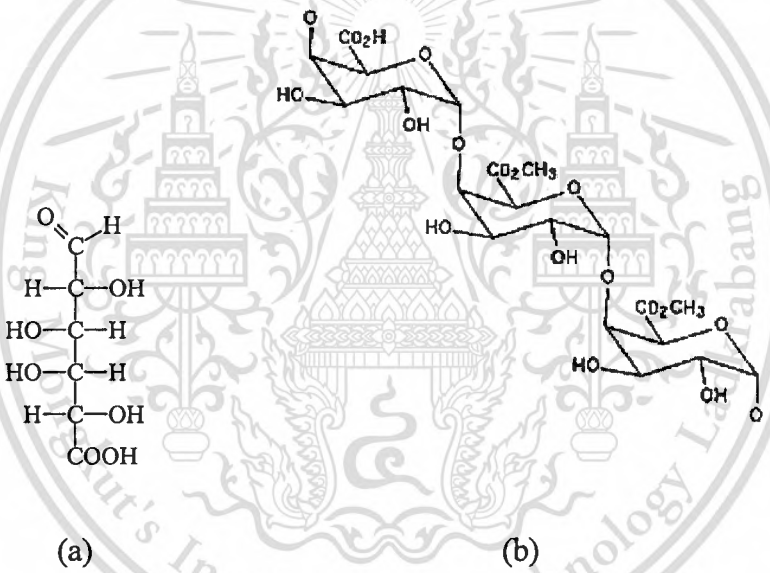


Figure 2.6 (a) galacturonic acid (b) Polygalacturonic acid (Pectin)

The capsular mucilage present in the seeds of *Ocimum canum Sims* has been isolate in 7% yield on seed weight basis, by cold water extraction and alcohol precipitation. The crude mucilage, an asbestos-like material, was found to contain on dry weight basis : N, nil; OCH_3, 2.14%; OAc, 4.01%; CCH_3, 0.38%; ash, 2.32%; lipids, free 24% and bound 5% and uronic acid by decarboxylation method, 8.15%.

2.4. Glycerol [7]

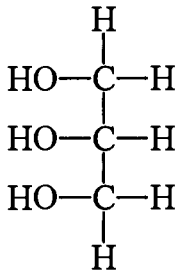


Figure 2.7 Structure of Glycerol. [7]

Glycerine is a somewhat viscous clear oily liquid, with a sweet taste. Synonyms of Glycerol include glycerin,1,2,3-trihydroxy propane, and 1,2,3 propanetriol. It is manufactured from petroleum, or from glycerides in fats, often as a by product in the production of soap. Glycerine is hydrophilic (water loving), and is used to keep products such as dried fruits from getting too dry, and to keep the liquid in soft fillings from escaping into crisp crusts, making them soggy. This emollient (moisturizing) effect also finds use in hand creams. It lowers the freezing point of water, and is used as an anti-freeze.

Glycerine is about ¾ as sweet as sugar, so it can reduce the need for sugar in products that require it for other reasons. In toothpaste it sweetens while keeping the paste from drying when the cap is left off. Glycerine is used in many skin creams and medicines, such as eye and ear drops, poison ivy creams, suppositories, and contraceptive jellies. Glycerine is used as an emollient (skin softener) in soaps.

2.5. Polyethylene glycol [8]

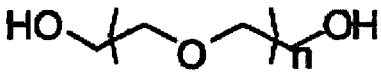


Figure 2.8 Structure of Polythylene glycol. [8]

Polyethylene glycol (PEG) is a family of long chain polymers made up of ethylene glycol subunits. The synonyms of polythylène glycol include PEG-n, PEG-6, PEG-8, PEG-12, PEG-40, PEG-150, etc. The number after the name indicates the approximate molecular weight of the chain (higher numbers mean longer chains). PEG is used as a thickener in many products. It is used in toothpaste to prevent bacteria from breaking down the pyrophosphates used to control tartar buildup. PEGs are often reacted with fatty acids to make detergents that have thickening and foam stabilizing properties. When chemically combined with fatty acids from coconut oil, it makes detergents like PEG-5 co-amide, which is used in shampoos as a surfactant, emulsifier, and foam stabilizer.

2.6. Biodegradable Polymers [9]

The definition of biodegradable polymer varies greatly scientists, manufacturers, and consumers. An American Society for Testing and Material (ASTM) definition of a biodegradable plastic involves a "plastic designed to undergo a significant change in its chemical structure under specific environmental conditions resulting in a loss of some properties. The degradation results come from the action of naturally-occurring micro-organism such as bacteria, fungi, and algae. Concerns with environmental fate will impose the additional requirement for properly designed biodegradable material of the complete "mineralization" or disappearance of the degradation products into CO_2, H_2O, CH_4, or biomass without the production of harmful intermediates. The time frame required for biodegradation will be mandated by the disposal method and conditions. The increase in composting and anaerobic bioreactor technology will produce specific environmental conditions and lead to specific requirements for biodegradable plastics packaging.

Plastics have come under attack in recent years due to their high visibility in the solid waste crisis. Non-degradable plastics packaging is blamed for shortening the life expectancy of commercial landfills, increasing the operational coat, contaminating the environment, and posing a treat to animal and marine life. Plastics account for approximately 15-17% of the $19 billion and food-packaging market, and it is predicted to increase to 50% by the year 2000. Nearly 10%of the plastics used in packaging are used as coasting on other materials, including paper. Increasing legislative and economic imperative and public perception of paper being natural and

plastics being "foreign" have led to the explosive drive to recycle plastic packaging or to make it "biodegradable."

Preferment, processability, and price comprise the typical requirements for biodegradable polymers. Performance is based on the ability to perform the necessary function during the service life as well as the disposal performance, biodegradability. Mechanical strength, processability, moisture and oxygen barrier properties, interactions with the product, printability, sterilizability, transparency, and inert to environmental exposures are a few of the properties necessary to evaluate for the service preferment. Due to the variety of applications, it is expected that numerous polymers or formulations will be required to best serve each application.

The biodegradation requirements will be determined by the disposal method and involve organisms, substrate exposure, and environmental factors. The availability of appropriate organisms, co-metabolism, and generation of appropriate enzymes for the biodegradation of the specific polymer need to be considered. The accessibility of the polymer, physical form, molecular weigh, branching, degree of crystallinity, and interactions with other components also play an important role. The environmental factors include temperature, oxygen availability, moisture level, pH, and nutrients. These requirements need to balance the degradability but hinder its application. The design should take into account any additives that may be harmful to the environment. These include cadmium-based pigments, heavy-metal stabilizers, and flame retardants.

The processability requirements of the material will be dictated by the manufacturing process, which is related to the thickness desired and the required properties. Melt processing techniques such as blown film extrusion, cylindering, injection molding, thermoforming, and the like will each require variations in melt viscosity and melt strength. The processing of the material can also affect the material properties as well as the biodegradation of the material.

2.7 Applications of Biodegradable Polymers [10]

Since the introduction of plastic films in the 1930s and 1940s for greenhouse coverings, fumigation and mulching, agricultural applications of polymers have grown at an enormous rate. All principal classes of polymers, i.e. plastics, coatings, elastomers, fibres and water-soluble polymers are presently utilized in applications which include the controlled release of pesticides and nutrients, soil conditioning, seed coatings, gel plantings and plant protection. However, degradable plastics are also of interest as agricultural mulches and agricultural planting containers. Ultimate biodegradability, as in composting, is also of some interest as it would permit degradable plastics to be combined with other biodegradable materials and converted into useful soil-improving materials.

2.7.1. Agricultural mulches

Mulches permit growers to use plastic films to help with plant growth and then photodegrade in the fields thereby avoiding the cost of removal. The plastic films are desirable because they conserve moisture, reduce weeds and increase soil temperatures, thus improving the rate of growth in plants. For example, a 6 ha melon farm reported a two- to three-fold increase in yield and ripening two weeks earlier as a result of using black polyethylene mulch. Elimination of weeds and avoidance of soil compaction by the use of mulch eliminates the need for cultivation, therefore root damage and stunting or killing of plants is further avoided. Fertilizer and water requirements are also reduced

Transparent polyethylene is more effective in trapping heat than black or smoke-grey films: soil temperatures may rise 5.5°C under clear films, as compared to 1.7–2.7°C under black films. Radiative heat loss at night, as the soil cools, is lessened by polymer films. In some cases, weed control has been reported because of solar heating of the polyethylene mulches. If left in place, however, conventional films can cause problems during harvesting or during cultivating operations the next year. Removal and disposal are costly and inconvenient. Therefore, interest in the development of biodegradable or photodegradable films with short service lifetimes has grown. Although a large number of polymer types could be designed for controlled degradation, only a few have been commercialized. Plastics used for this

purpose usually contain light-sensitizing additives which cause the materials to undergo photodegradation.

The plastics used for mulch films are generally low density polyethylenes, poly(vinyl chloride), polybutylene or copolymers of ethylene with vinyl acetate. Biodegradable films based on starch with poly(vinyl alcohol) poly(ethylene-*co*-acrylic acid) and poly(vinyl chloride) have been developed in the USDA laboratories. Polylactone and poly(vinyl alcohol) films are readily degraded by soil microorganisms, whereas the addition of iron or calcium accelerated the breakdown of polyethylene. Degradable mulches should break down into small brittle pieces which pass through harvesting machinery without difficulty and do not interfere with subsequent planting.

2.7.2. Controlled release of agricultural chemicals

Controlled release (CR) is a method by which biologically active chemicals are made available to a target species at a specified rate and for a predetermined time. The polymer serves primarily to control the rate of delivery, mobility, and period of effectiveness of the chemical component. The principal advantage of CR formulations is that less chemicals are used for a given time period, thus lowering the impact on non target species and limiting leaching, volatilization, and degradation. The macromolecular nature of polymers is the key to limiting chemical losses by these processes.

Starch, cellulose, chitin, alginic acid, and lignin are among the natural polymers used in CR systems. These have the advantages of being abundant, relatively inexpensive, and biodegradable. Although they possess functionality for derivatization, they have the one significant disadvantage of being insoluble in standard solvents suitable for encapsulation, dispersion, and formulation. Systems have been developed that overcome the solvent problem by *in situ* encapsulation, whereby gelantinized starch containing a chosen pesticide is cross-linked by adding calcium chloride or boric acid, or by xanthanation followed by oxidation. The pesticide, as a result, is entrapped within the granular particles formed.

2.7.3. Agricultural planting containers

A small niche for degradable plastics is the use of polycaprolactone for small agricultural planting containers. Although this is a small volume application for degradable plastics, it is presented here because it is one of the few applications in which the polymer used is biodegradable within a reasonable period of time. These polycaprolactone planting containers have been used for automated machine planting of tree seedlings. Within six months in the soil, the polycaprolactone was found to undergo significant biodegradation, resulting in 48% weight loss, with 95% weight loss occuring in a year.

2.7.4. Packaging

Physical characteristics of packaging polymers are greatly influenced by the chemical structure, molecular weight, crystallinity and processing conditions of the polymers used. The physical characteristics required in packaging depend on what item will be packaged as well as the environment in which the package will be stored. Items which must be kept frozen for a period of time require special packaging. Food items require more stringent packaging requirements than nonperishable goods.

The challenge in the development of biodegradable packaging will be to combine polymers which are truly biodegradable into a laminate film or a film blend which has properties as good as those found in synthetic laminates. For food applications, for example, it may be possible to coat food items with pullulan which has very low oxygen permeability and is edible and to utilize PHBV as an outer packaging which has good flexibility and is a moisture barrier. A film blend of pullulan and PHBV can also be produced since both polymers can be melting blended under conditions where sufficient moisture is maintained during processing. The addition of pullulan to PHBV may reduce oxygen permeability and increase biodegradability of the blend due to the increased surface area of PHBV exposed following the rapid removal of pullulan due to its water solubility.

Several polysaccharide-based biopolymers are being used as possible coating materials or packaging films. They include starch, pullulan and chitosan. The degradation of synthetic polymer films can be accelerated by incorporating starch as a

filler. LDPE blends with up to 10% corn starch were produced using conventional techniques and were made into bags for groceries or rubbish.

2.8 Research Methodology

I. M. Thakore, S. Desai, B. D. Sarawade and S. Devi [11] study on corn starch was modified by cross-linking with epichlorohydrin and plasticizer glycerol. X-ray diffraction studies showed that relative crystallinity of the native and cross-linked starch were similar and were not affected by cross-linking. Different films were prepared by blending corn starch, cross-linked starch or glycerol modified starch in LDPE. The mechanical properties of the films were studied for tensile strength, elongation, melt flow index, and burst strength. The properties of the blend films were compared with LDPE films. It was observed that with the blending of 7.5% native starch, there was a decrease in tensile strength, elongation and melt flow index but burst strength increased. The tensile strength, elongation and melt flow index of the films containing cross-linked starch was considerably higher than those containing native starch but the burst strength showed a reverse trend. For native starch and cross-linked starch modified with glycerol, the elongation and melt flow index of the films increased but burst strength decreased. Surface scanning of the blend films were done by scanning electron microscope. Film containing cross-linked starch/glycerol modified starch in the blend was observed to be smoother than the native starch blend films.

E.M. Nakamura, L. Cordi, G.S.G. Almeida, N. Duran and L.H.I. Mei [12] study on investigated the incorporation of different starches, such as native, adipate, acetylated and cassava starch, in low-density polyethylene matrix (LDPE) to verify the possibility to obtain partially biodegradable product with the aim to decrease the plastics wastes in the environment. The starches were mixed to the LDPE using a high shearing mixer to guarantee the homogeneity of the formulations. The characterization of those compounds was done using mechanical and morphological analysis and biodegradation in activated sludge. The results indicated that the increase of the starch amount into the olefin matrix was responsible for the reduction on mechanical properties of the products, if compared with pure LDPE. It was observed by SEM that the activated sludge is a promising method to follow biodegradation.

F. J. Rodriguez-Gonzalez, B. A. Ramsay and B. D. Favis [13] study under the particular one-step processing conditions used it is possible to develop continuous TPS (highly interconnected) and co-continuous polymer/TPS blend extruded ribbon which possess a high elongation at break, modulus and strength in the machine direction. The PE/TPS (55:45) blend prepared with TPS containing 36% glycerol maintains 94% of the elongation at break and 76% of the modulus of polyethylene. At a composition level of 71:29 PE/TPS for the same glycerol content, the blend retains 96% of the elongation at break and 100% of the modulus of polyethylene. These excellent properties are achieved in the absence of any interfacial modifier and despite the high levels of immiscibility in the polar–nonpolar TPS–PE system. The 55:45 blends possesses a 100% continuous or fully interconnected TPS morphology, as measured by hydrolytic extraction. This highly continuous TPS configuration within the blend should enhance its potential for environmental biodegradation. The elongation at break in the cross direction of these materials, although lower than the machine direction properties, also demonstrates ductility at high TPS concentrations. At a glycerol content of 36% in the TPS, the blends demonstrate only very low levels of sensitivity to moisture. A high degree of transparency is maintained over the entire concentration range due to the similar refractive indices of PE and TPS and the virtual absence of interfacial micro voiding.

Y. J. Kim, Y. M. Lee, H. M. Lee and O. O. Park [14] study on an investigation of the biodegradable polymers based on starch has been accomplished. Starch needs to be modified before blending with low density polyethylene (LDPE) because of its hydrophilicity. Three kinds of modification methods were applied to obtain the compatibility to starch LDPE blends. The first one was the esterification of the hydroxyl group of starch with acetic anhydride. The second was the copolymerization of acrylonitrile and styrene onto starch. The last was the esterification reaction of the hydroxyl group of starch with three kinds of ionomers. The tensile strength, % elongation, Young's modulus, and shear viscosities of blends of the modified starch and LDPE were examined. Ionomer-treated starch showed better compatibility with LDPE to give better performances than other blends.

D. Bikiaris, J. Prinos, K. Koutsopoulos, N. Vouroutzis, E. Pavlidou, B. N. Frangi and C. Panayiotou [15] study a series of polyethylene/plasticized starch blends were prepared using a poly(ethylene-g-maleic anhydride) copolymer as a reactive compatibilizer. Uncompatibilized blends were also prepared for comparison purposes. The prepared blends were studied using mechanical properties measurements and SEM microscopy to determine their morphology. The blends were also exposed to activated sludge to determine their biodegradability.

H. Dave, P.V.C. Rao and J.D. Desai [16] study on starch-containing low density polyethylene films tested for biodegradation, films containing 30% starch showed maximum biodegradation leading to a loss of 6.3% in weight and 84.5% starch upon burial in a soil ± compost mixture for 48 weeks. Biodegradation of this film was accelerated by incubation with an acclimatized culture in a shake-ask showing 11.2% and 68.9% loss of weight and starch respectively, after only 6 weeks. FTIR and 13C NMR analysis revealed that the loss of starch in biodegraded films is accompanied by structural changes in polyethylene in terms of reduction in the peak area of polyethylene backbone and short chain branches.

I. Jakubowicz [17] study on thermo-oxidative degradation of polyethylene films containing pro-oxidant has been studied at three temperatures that normally occur during composting conditions. Besides temperature, oxygen concentration was also varied. After various periods, the effects of thermo-oxidation were evaluated by measurements of molecular mass of the materials. It is shown that while temperature is the most important factor influencing the rate of thermo-oxidative degradation of the materials, oxygen concentration is of negligible importance. The investigation has also shown that when the material is degraded into low molecular mass products, it is bioassimilated. The rate of aerobic biodegradation of the oxidation products was evaluated under controlled composting conditions using measurements of produced carbon dioxide. The degree of bioassimilation in our case was about 60%, and still increasing, after 180 days.

R. Chandra and Renu Rustgi [18] study on environmental threats restrict the use of nondegradable polymers and encourage the development and use of degradable plastics. In order to obtain a cost-effective biodegradable plastic, starch-filled polyethylene (PE) is still the best alternative. Starch and PE blend is incompatible at

the molecular level and often leads to poor performance. In order to overcome this drawback, either PE or starch should be modified. The aim of this study was to modified linear low-density polyethylene (LLDPE) and blends it with starch. Maleic anhydride (MA) was grafted onto LLDPE in xylene using dicumyl peroxide (DCP) as an initiator. Corn starch in varying concentrations (between 10 and 60%) was blended with MA-g-LLDPE in a torque rheometer. The same blend compositions of nonfunctional LLDPE with the starch were prepared for comparative studies. The torque and totalized torque generated during blending are reported as a function of starch content. Torque decreased with increasing starch content for the compositions from 10 to 50% and increased for 60% starch content. Work energy decreased for all the compositions of blends except for 60% starch content. Tensile strength and modulus increased and percentage elongation decreased as the starch content increased in the blends. Water absorption of the blends increased with an increase in starch content. The biodegradability of MA-g-LLDPE/starch blends have been studied in two biotic environments: (1) soil environment over a period of 6 months; (2) mixed fungi inoculum (Aspergillus niger, Penicillium funiculosum, Chaetomium globosum, Gliocladium virens and Pullularia pullulans) for 28 days. The samples containing more than 30% starch content supported heavy fungus growth. Blends exposed to a soil environment degraded more than in fungi alone. Any changes in the various properties of the MA-g-LLDPE/starch before and after degradation were monitored using FTIR spectroscopy, weight loss, a scanning electron microscope (SEM) for surface morphology, a differential scanning calorimeter (DSC) for crystallinity and a thermogravimetric analyzer (TGA) for rapid determination of starch content. Percentage crystallinity decreased as the starch content increased and biodegradation resulted in an increase of crystallinity in MA-g-LLDPE/starch blends.

Chapter 3

Experimental

3.1. Materials

3.1.1 Low Density Polyethylene (LDPE) Code LD1905F from Thai Polyethylene CO, LTD.

3.1.2 Hairy Basil seed (*Ocimum canum Sims*) from Raithip CO, LTD.

3.1.3 Distilled water

3.1.4 99.5% Glycerol, Univar reagent, analytical grade B/NO. AH501173

3.1.5 Polyethylene glycol (PEG) M.W. 400 from Fluka

3.2 Apparatus

3.2.1 An enameled basin

3.2.2 Aluminum tray

3.2.3 Cloth filter

3.2.4 National MX-110PN electrical blender with container

3.2.5 WTC BINDIR-7200 tuttlingen Oven

3.2.6 Retsch ZM100 grinder with 0.08 mm sieve

3.2.7 Lab Tech engineering CO, LTD-LRM110 Two-roll mill

3.2.8 Desiccators

3.2.9 Glass rod

3.2.10 BOSCO BG2305 granulators with screen pore size 6 mm. diameter.

3.2.11 Thermo Haake Polydrive Extrusion Blown-Film machinery

3.2.12 LLOYD Universal Testing Machine LR5K

3.2.13 Fourier transforms infrared spectrometer (FT-IR), Spectrum GX, PerkinElmer

3.2.14 Thermogravimetric analyser (TGA), Pyris 1 TGA, Perkin Elmer

3.2.15 CEAST-6841 Melt Flow Rate Tester

3.2.16 LEO 1455 VP Scanning Electron Microscope Analyser

3.3 Preparation of mucilage powder

1. Put 100 grams of Hairy Basil seed into an enameled basin.

Figure 3.1 Weighing of Hairy Basil seed

2. Pour 3.5 liters of distilled water into an enameled basin, allow it to stand for 1 day.

(a)

(b)

Figure 3.2 (a) Adding distilled water into Hairy Basil seed

(b) Hairy Basil seed swollen in distilled water.

3. Swollen seed was pour into an electrical blender, stirred for 1 minute, for extraction mucilage gel from its black seed.

Figure 3.3 Pouring the swollen-seed into an electrical blender.

4. Filter black seed from the mucilage gel by pouring gel solution on cloth filter and then squeeze, as shown in the figure below.

(a) (b)

Figure 3.4 (a) Pouring the gel solution into the white cloth filter

(b) Pour cloudy mucilage gel into aluminum tray.

5. Dry this mucilage gel in the oven at 50 °C for 1 day.

(a) (b) (c)

Figure 3.5 (a) Mucilage gel in the tray, (b) mucilage gel in the oven, and (c) the oven.

6. After drying, dry mucilage sheet was grinded into powder by using 0.08 mm Retsch grinder sieve.

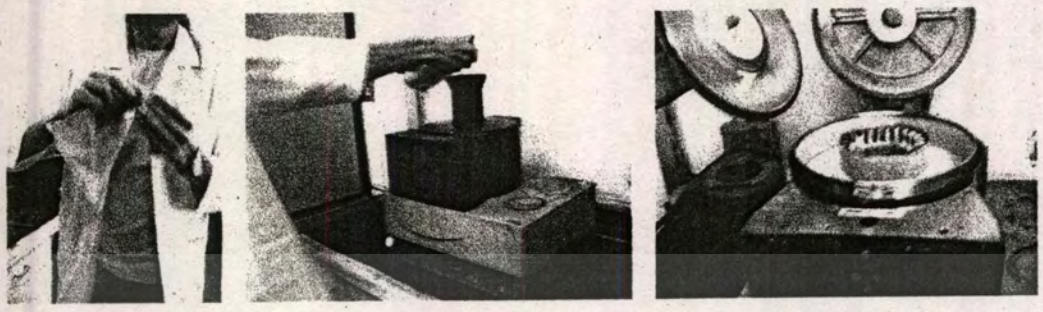


Figure 3.6 (a) Tear dry mucilage sheet into smaller size (b) Put mucilage sheet into the Retsch grinder, and (c) Mucilage powder was put in the Retsch grinder.

7. Keep mucilage powder in desiccators.

3.4 Preparation of blended LDPE

1. Weigh glycerol or polyethylene glycol (used as plasticizer of mucilage powder) for each formula, as shown in Table 3.1.

Figure 3.7 Weigh of plasticizer.

2. Weigh mucilage powder as shown in Table 3.1 for each formula.

Figure 3.8 Weigh of mucilage powder.

3. Gradually put mucilage powder in glycerol or polyethylene glycol and gradually stirred by hand until the whole mucilage powder totally wet and became pastry.

Figure 3.9 Mixing the mucilage powder with plasticizer.

4. Weigh LDPE pellets as shown in Table 3.1 for each formula.

Figure 3.10 Weigh of LDPE.

5. Warm up two-roll mill at 125°C for 10 minutes and then put LDPE pellets.

Figure 3.11 Pour LDPE on the surface of two-roll mill.

6. After LDPE pellets already melt and stick on the rollers, add the pastry of mucilage and plasticizer to molten LDPE and knead for 15 minutes.

(a) (b)

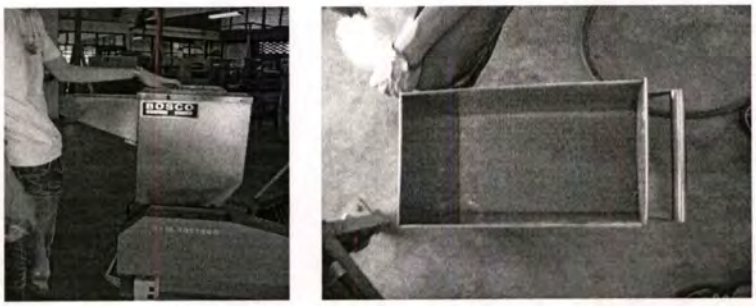
Figure 3.12 (a) Molten sheet of mixed LDPE on the surface of two-roll mill

(b) Cut and fold technique.

7. After mixing 15 minutes, take out the blended plastics and cut into small lumps and then leave it for 15 minutes cooling

Figure 3.13 Lump of LDPE blend with mucilage powder.

8. Reduce size of blend plastic lumps by using granulator with 0.6 mm. sieve.



(a) (b)

Figure 3.14 (a) The granulator (b) LDPE/mucilage pellet ready to use.

Table 3.1: Variation of mucilage powder, amount of glycerol or polyethylene glycol and LDPE pellets.

Formula	Glycerol (g)	PEG (g)	*Mucilage* powder (g)	LDPE pellets (g)
1	10	-	10	300
2	-	10	10	300
3	-	20	20	300
4	-	15	30	300
5	-	15	20	300
6	-	15	15	300

3.5 Preparation of blended LDPE Film

1. Turn on the machine and set temperatures required (conditions shown in table 3.2.)

2. Put the blended LDPE into hopper.

3. Pull up the blended LDPE melt from O-ring up to the roller.

4. Keep up film product.

Table 3.2: The production film conditions of blown film extrusion machine.

Variables	Conditions
Zone 1 barrel temperatures	130°C
Zone 2 barrel temperatures	140 °C
Zone 3 barrel temperatures	150 °C
Zone 4 barrel temperatures	160 °C
Screw speed	100 rpm
Take up roll speed	4 mm/s

3.6 Characterization of LDPE/mucilage film

3.6.1. Melt Flow index

LDPE/mucilage pellets were put into 190°C cylinder and applied 2.16 kg load. Then, collect the melt LDPE/mucilage which passed through the cylinder within 10 minutes for weighing. The value of MFI is proportional to viscosity of the mixture.

3.6.2. Thermogravimetric Analysis (TGA)

Thermal property was performed by using thermogravimetric analyzer (TGA). This technique is use to determine the decomposition temperature of mucilage powder, polyethylene glycol or glycerol and LDPE/mucilage film by putting sample in a chamber inside the furnace. TGA was performed under nitrogen flow using temperature range of 50 – 800°C at heating rate of 10°C/min.

3.6.3. Functional group analysis by Fourier Transform Infrared Spectroscopy technique (FT-IR).

The Fourier transformed infrared spectra (FT-IR) of LDPE/mucilage film were recorded using FT-IR spectrometer, Spectrum GX, Perkin Elmer. The characteristics of the LDPE/mucilage film were obtained using a transmission mode of FTIR. The samples were scanned from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}.

3.6.4 Measurement of Water Uptake

Sample films were allowed to swell in excess water (pH 7.0) at room temperature. At time interval, sample was removed from water and weighed after excess on the surfaces was blotted. The swelling ratios or water uptake of sample film was calculated from the following equation:

$$WU\ (\%) = \frac{\text{Wet weight} - \text{Dry weight}}{\text{Dry weight}} \times 100$$

3.6.5 Measurement of Tensile and Elongation at break

An ASTM D 882-02 Universal Testing Instrument was used to determine tensile strength, elongation at break and modulus. The samples were 38 mm long and 8.0 mm wide strips. The initial grip separation was 50 mm and the strain rate was 12.5 mm/min.

3.6.6 Soil Burial Test

LDPE/mucilage film samples were embedded under the soil for 4 months and the samples were collected for measuring weight lost every month. After 4 months, samples were investigated by microscopy to check surface deterioration.

Chapter 4

Results and Discussion

The preparation of blended LDPE was done by separated mucilage from *Ocimum canum Sims* and mixed it with plasticizer (PEG or glycerol), varied the weight ratio of mucilage and plasticizer, then prepared film by blown film extrusion machine. The blended LDPE could be characterized by MFI, TGA, FT-IR, water uptake, tensile, elongation, modulus and soil burial.

4.1. Melt Flow index

Table 4.1: The results of each formula.

Formula	G (g)	P (g)	*Mucilage* powder (g)	LDPE pellets (g)	*Mucilage* powder in 100 g LDPE	Blown film preparation results	MFI (g/ 10mins)
1	10	-	10	300	3.33	X	ND
2	-	10	10	300	3.33	√	4.285
3	-	20	20	300	6.67	X	5.50
4	-	15	30	300	10.00	X	4.26
5	-	15	20	300	6.67	X	4.68
6	-	15	15	300	5.00	√	5.14

Note: G = Glycerol P = Polyethylene glycol 400

X = can not prepared film √ = can prepared film

ND = no data

Among 6 formulae, only formula 2 (mucilage = 3.33%) and formula 6 (mucilage = 5.0%) could be prepared by blown film process. This means that the optimum mucilage powder should be 5.00%

It was found that MFI has no effect to the process. Because formula 2 and 6 which could be prepared blown film have different in MFI, the formula 3(MFI = 4.26 g / 10mins) which has closely MFI of formula 2(MFI = 4.29) could not prepare blown film.

When consider the ratio of plasticizer (polyethylene glycol) with mucilage powder found that appropriate ratio for preparation of blown film is 1:1 and mucilage powder could not more than 5%.

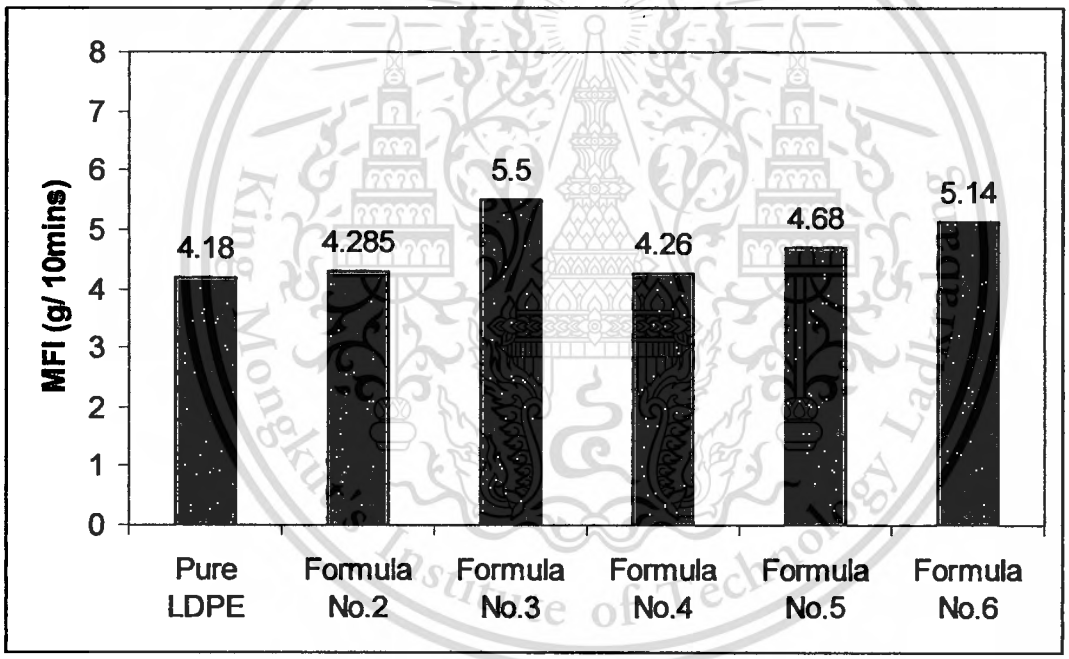


Figure 4.1: Relation between pure LDPE and formula 2-6 with MFI (g / 10mins)

Formula 2, 6 and 3 which have the same ratio of PEG to mucilage powder (1:1) but different quantity were shown drastically change, in MFI. Formula 3 containing 20 g. of mucilage powder exhibited the highest MFI. The results indicated that the more plasticizer could be enhanced more free volume in LDPE matrix.

Comparison of sample 4, 5, and 6 which contain same amount of PEG but different amount of mucilage powder should be noted that the more content of

mucilage powder (formula 4) indicated the lowest MFI. This means that addition of mucilage powder retards the mobility of molten LDPE chain leading to high viscosity and low MFI.

To compare the efficiency of PEG on MFI, it should be considered formula 2, 4, and 3. MFI of LDPE composites obviously increased from 4.28 to 5.5 g/ 10 mins regardless of mucilage content.

Incorporation of mucilage powder is increased viscosity of the blended LDPE while the addition of PEG is reduced viscosity.

4.2. Thermogravimetric Analysis (TGA)

The decomposition temperature of the low density polyethylene and mucilage powder was investigated by TGA under nitrogen gas. The amount of organic matter could be determined from the weight loss of the substances in the compound.

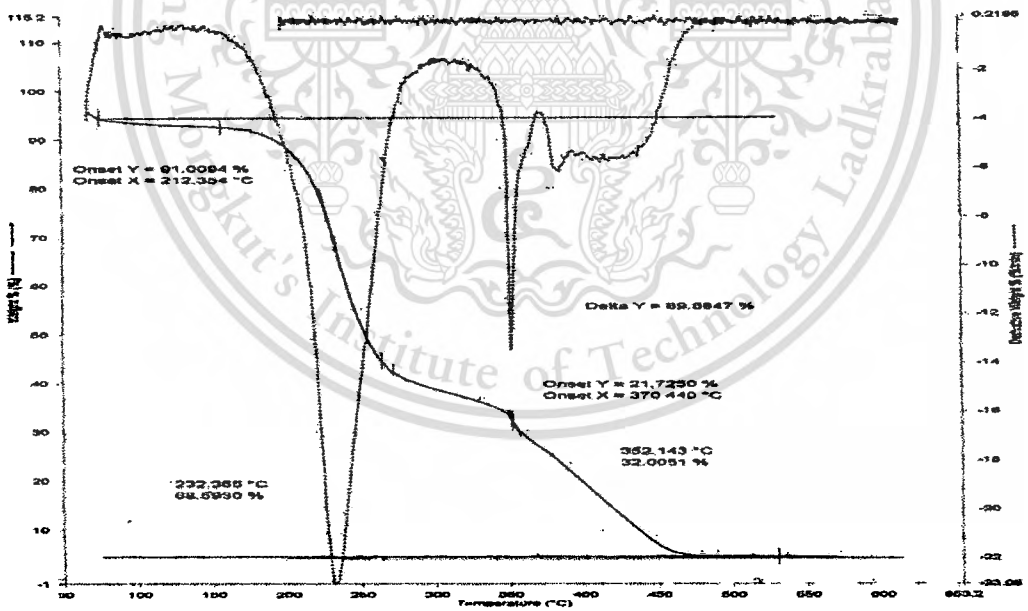


Figure 4.2: TGA thermogram of mucilage powder

Figure 4.2 presents the TGA diagram resulting from mucilage powder. The pyrolysis of mucilage powder takes place in 3 steps. The first one is in the 232°C and corresponds to degradation of glycosyl units [19]. The second one at 350°C and the

and the third one from 370-430°C are attributed to the degradation of methyl ester and carboxyl groups, respectively.

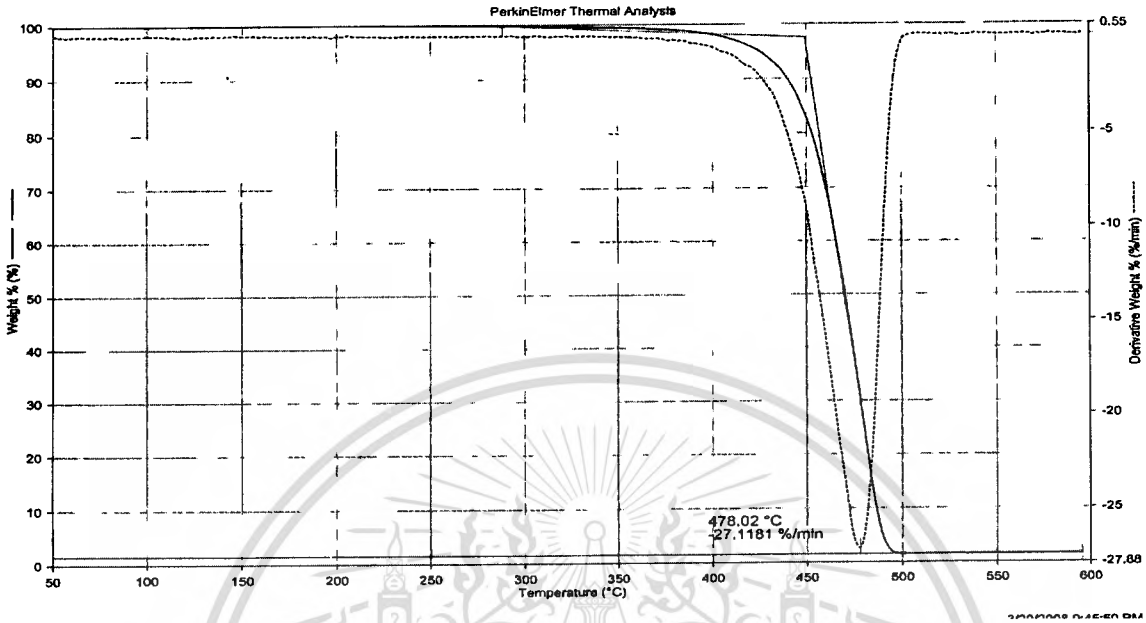


Figure 4.3: TGA thermogram of LDPE.

The decomposition temperature of LDPE is about 478°C as shown in figure 4.3.

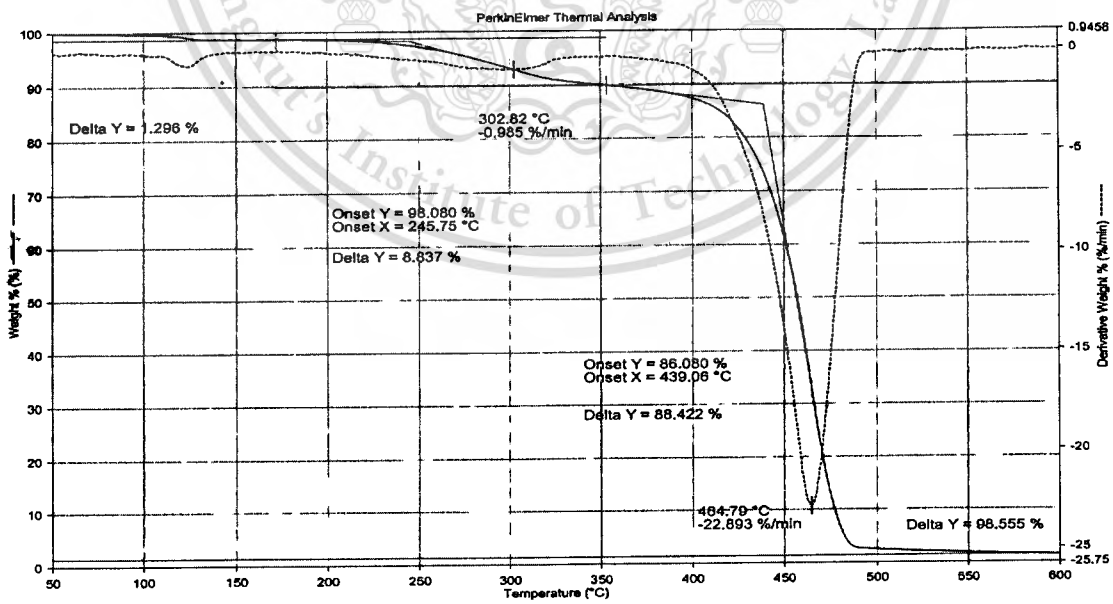


Figure 4.4: TGA thermogram of formula 4 (15 PEG:30 mucilage).

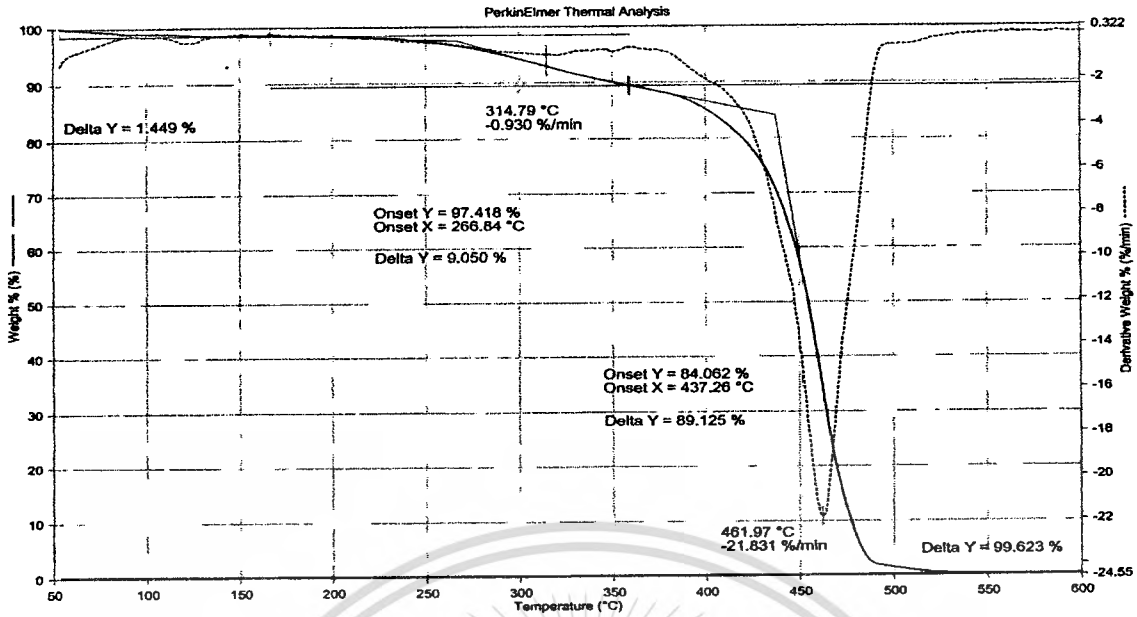


Figure 4.5: TGA thermogram of formula 5 (15 PEG:20 mucilage).

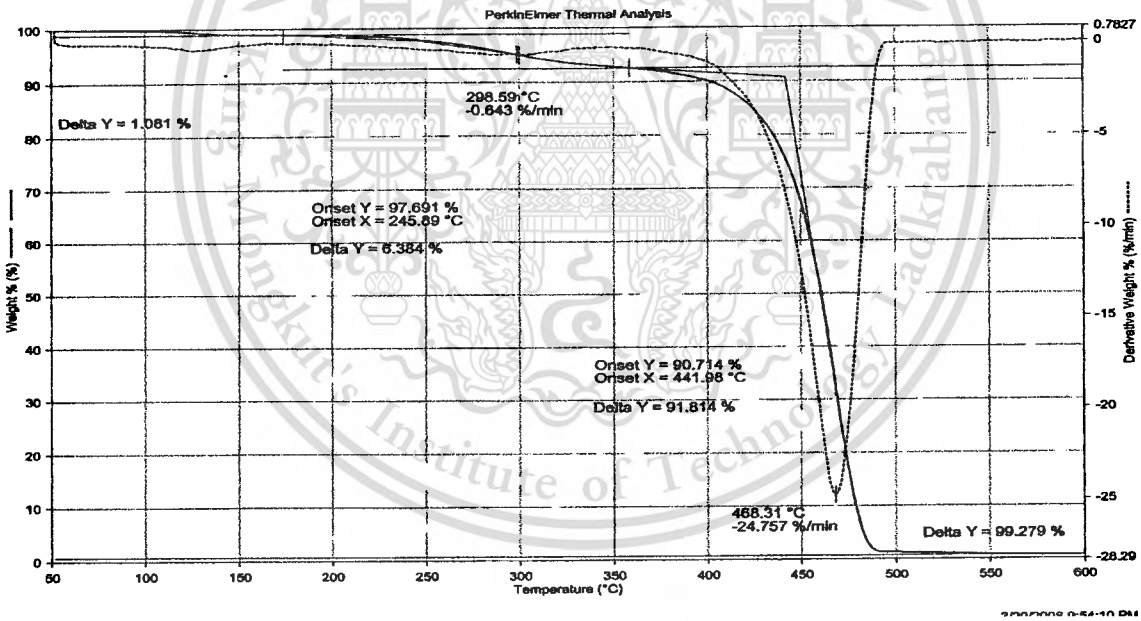


Figure 4.6: TGA thermogram of formula 6 (15 PEG:15 mucilage).

Figure 4.4-4.6 are presented the TGA diagram resulting from LDPE:PEG:mucilage in formula 4. Mass loss around 300°C should be ascribed to PEG loss. A significant mass loss occurs with the temperature range of 460-480°C due to the pyrolysis of LDPE. Therefore with increasing mucilage powder content in blended LDPE, the peaks of derivative weight will having lower temperature. This

result indicates that mucilage powder probably interfere the interaction of LDPE chain.

Table 4.2: The conclusion of decomposition temperature by TGA analysis

Materials	Decomposition temperature (°C)	
	1st	2nd
Mucilage powder	232	352
LDPE	478	-
Formula 4(15PEG-30mucilage)	302	464
Formula 5(15PEG-20mucilage)	314	461
Formula 6(15PEG-15mucilage)	298	468

Figure4.2-4.6 are shown the TGA thermograms of the starting mucilage powder, low density polyethylene, LDPE with 15PEG-10mucilage, 15PEG-15mucilage, 15PEG-20mucilage, and 15PEG-30mucilage respectively. The thermogram of mucilage powder was shown the highest weight loss at 232°C. For the low density polyethylene, the highest weight loss was at 478°C. The other mixture thermograms are shown of low density polyethylene with mucilage powder and polyethylene glycol, the two highest weight losses were around 300°C and 470°C. At around 470°C it is shown the weight loss of low density polyethylene. The higher the amount of mucilage powder, the lower temperature is required to decompose.

4.3. Functional group analysis by Fourier Transform Infrared Spectroscopy technique (FT-IR).

Typical chemical characteristics of both organic and inorganic materials could be studied by FTIR spectra analysis, and also be an appropriate technique to study the mucilage powder and polyethylene glycol interaction with low density polyethylene, because of a shift in υ_{OH} vibration.

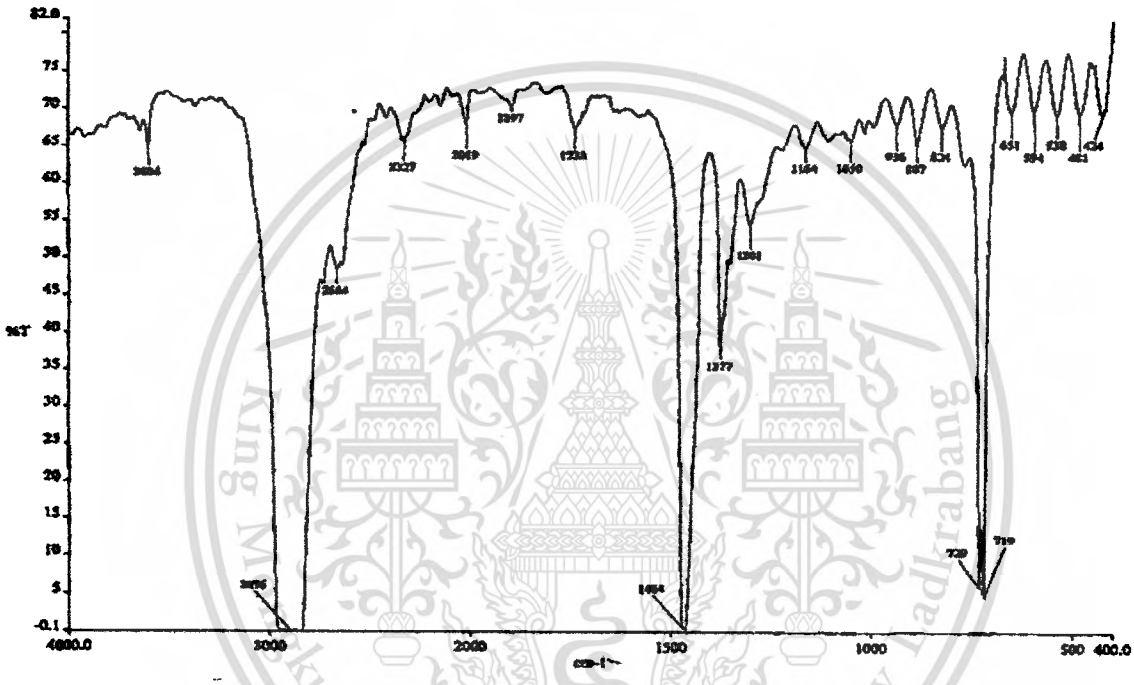


Figure 4.7: Spectrum of low density polyethylene

Figure 4.7 shows υ_{CH2} for CH_2 stretching with the range at 2890 cm^{-1}, υ_{CH2} for CH_2 bending ~1485-1445 cm^{-1}, and υ_{CH2} for CH_2 rocking ~750-720 cm^{-1}

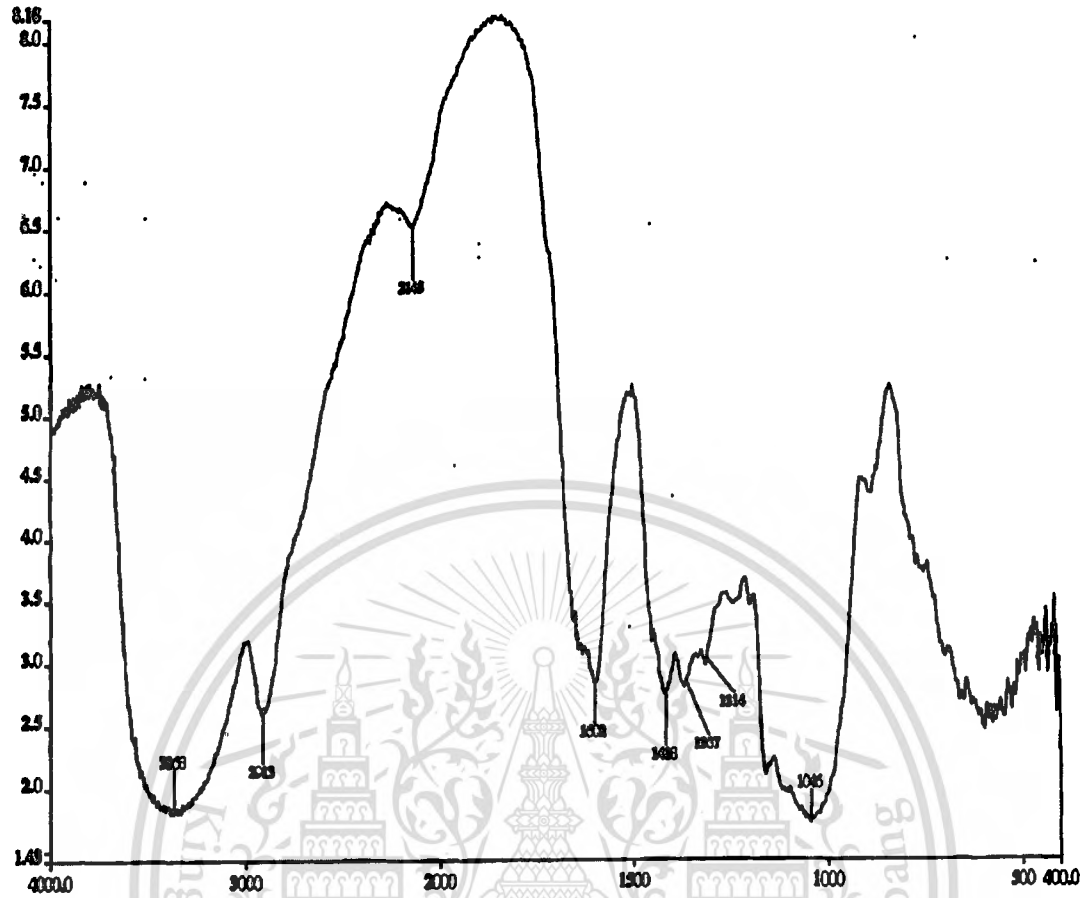


Figure 4.8: Spectrum of mucilage powder

Figure 4.8 shows υ_{OH} for –OH ~ 3400-3200 cm^{-1}, υ_{CO} of C=O stretching ~ 1600 cm^{-1}, and υ_{CO} for C-O stretching ~1000 cm^{-1}

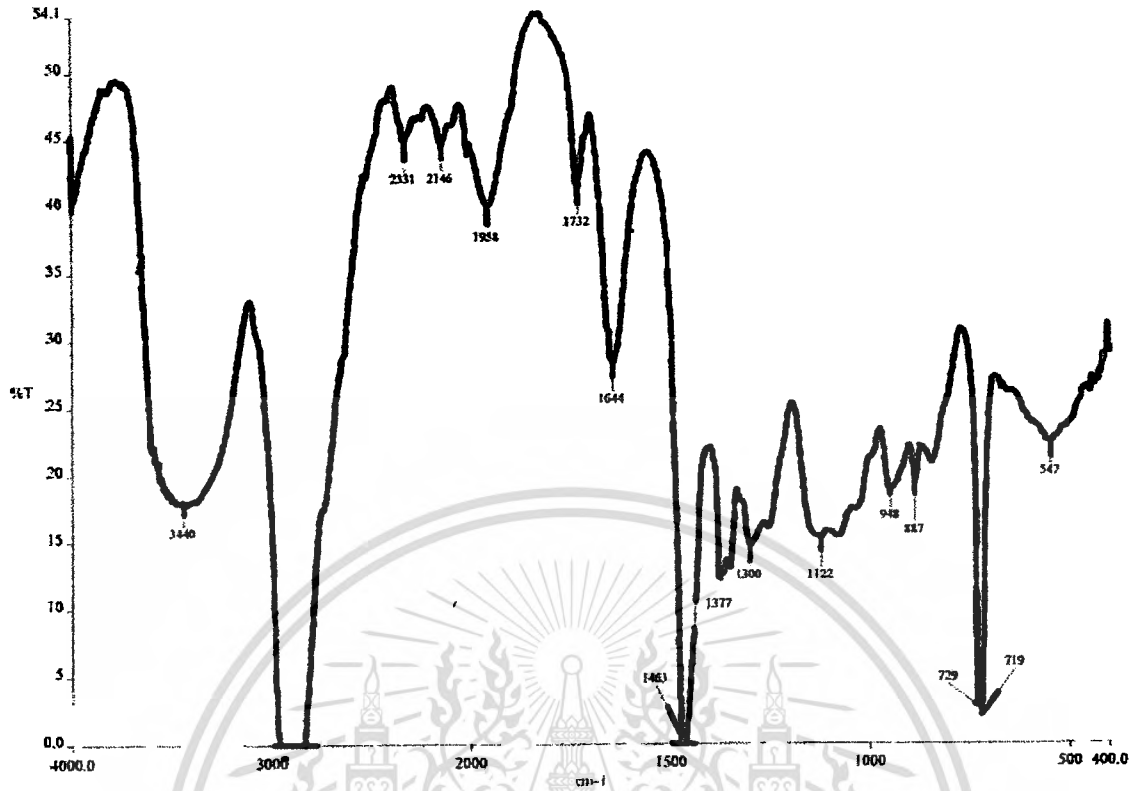


Figure 4.9: Spectrum of polyethylene glycol with LDPE

The spectrum of low density polyethylene, the spectrum of mucilage powder, and the spectrum of polyethylene glycol are shown in Fig.4.7 to Fig. 4.9, respectively. The result indicated that the peak of low density polyethylene is shown the peak vibration band of the hydrocarbon as follows; υ_{CH2} for CH_2 stretching with the range at 2890 cm^{-1}, υ_{CH2} for CH_2 bending ~1485-1445 cm^{-1} , and υ_{CH2} for CH_2 rocking ~750-720 cm^{-1}. For the mucilage powder major peaks at υ_{OH} for –OH ~ 3400-3200 cm^{-1}, υ_{CO} of C=O stretching ~ 1600 cm^{-1} , and υ_{CO} for C-O stretching ~1000 cm^{-1} .

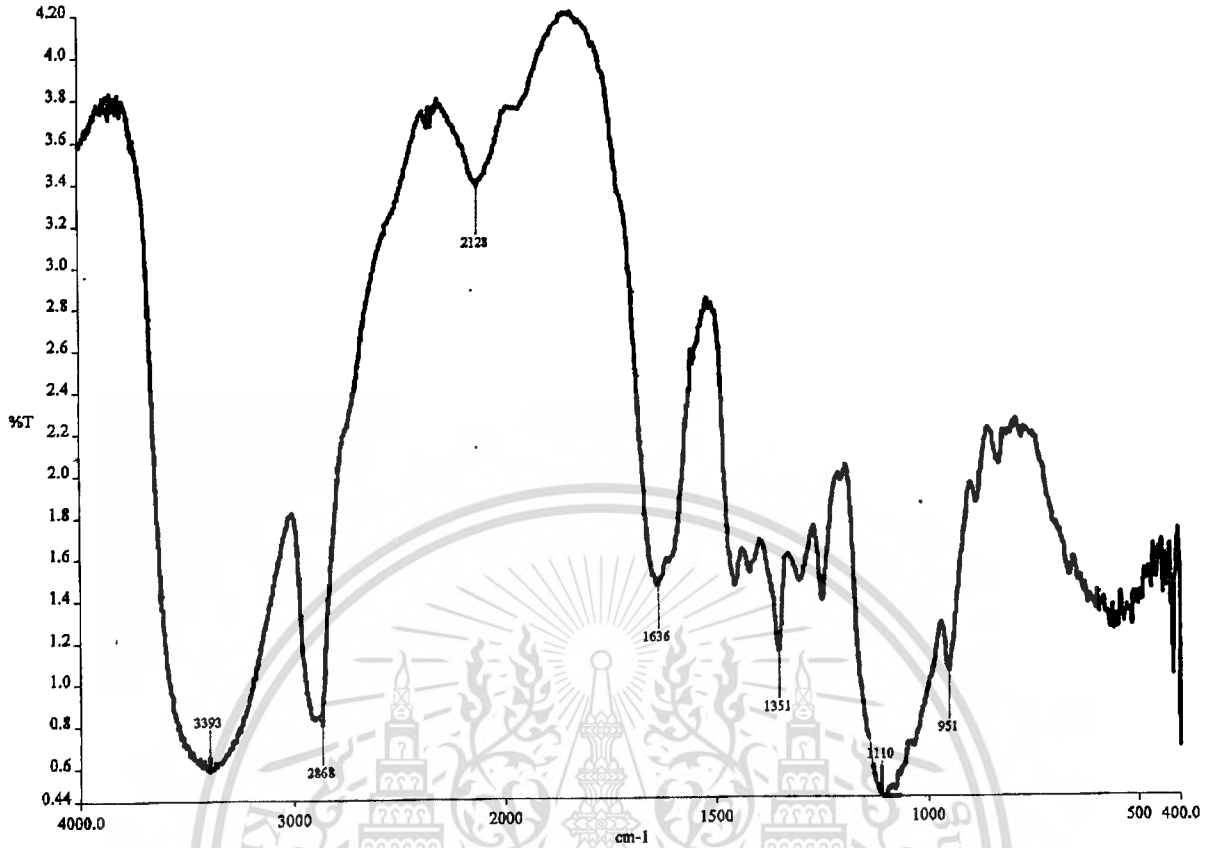


Figure 4.10: Spectrum of mucilage powder with polyethylene glycol

Figure 4.10 shows υ_{OH} for –OH ~ 3400-3200 cm^{-1}, υ_{CO} of C=O stretching ~ 1600 cm^{-1}, and υ_{CO} for C-O stretching ~1000 cm^{-1}

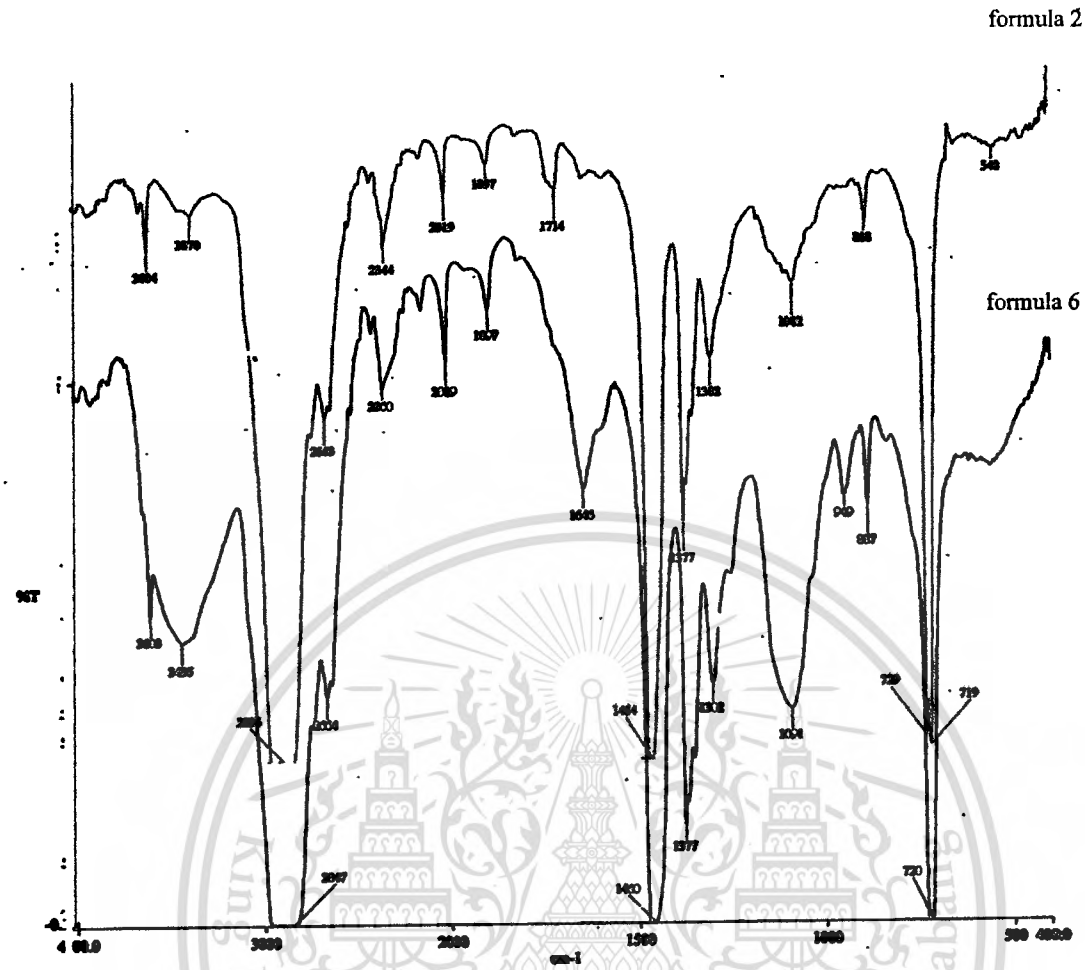


Figure 4.11: Spectrum of low density polyethylene formula2 (mucilage = 3.33%) compare with formula 6 (mucilage = 5.0%)

Figure 4.11, at 3400-3300 cm^{-1} confirm the amount of hydroxyl group for formula6 is higher also at ~1000 cm^{-1} confirm the amount of ether group for formula6 is higher.

When compare formula 2 and 6, the formula 2 has more clearly absorption frequency at 3435 cm^{-1} than formula 6. It means that formula 2 has higher amount of polyethylene glycol.

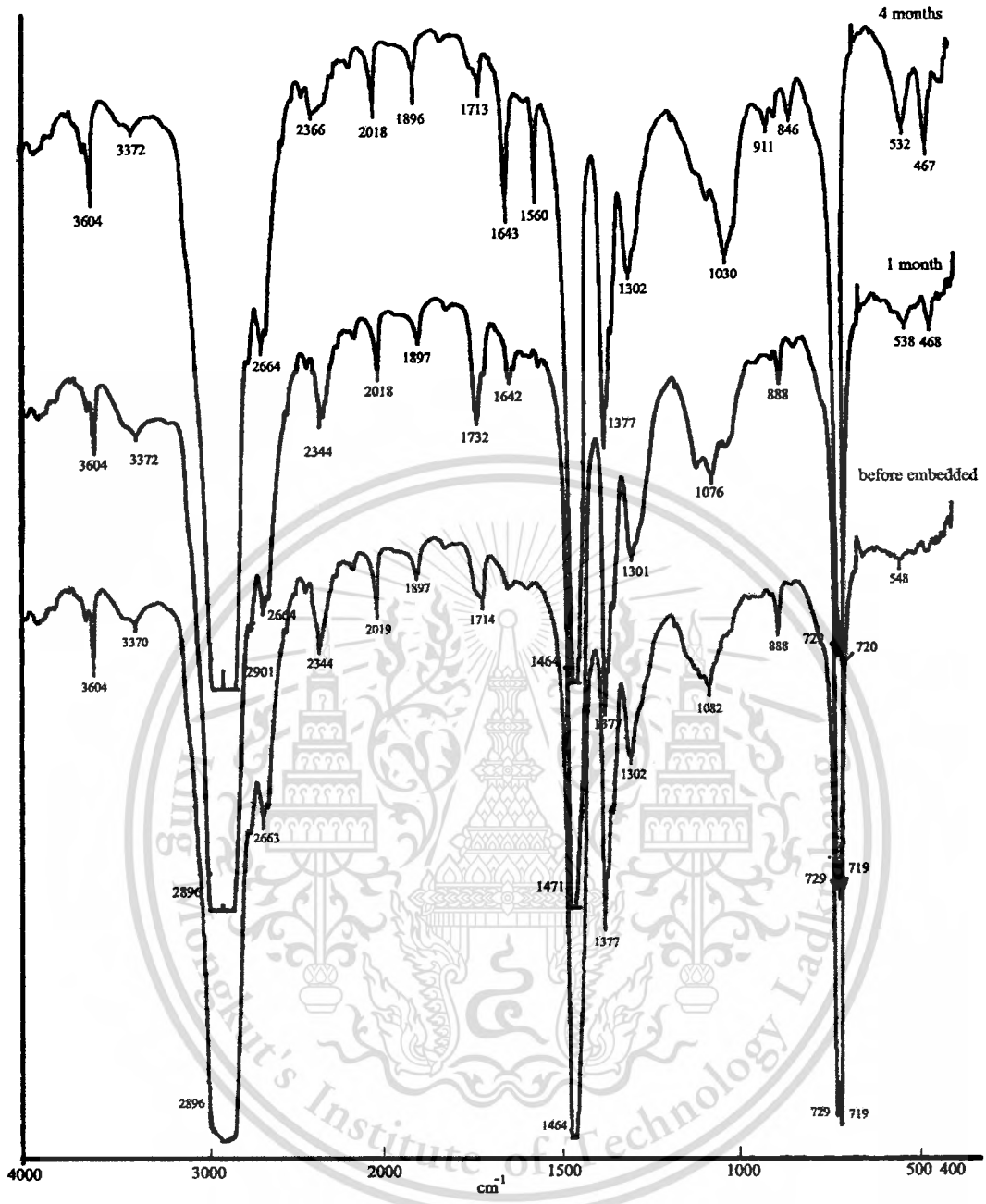


Figure 4.12: The comparison of FT-IR spectra for formula2 (mucilage = 3.33%) before and after burial test.

Fig.4.12 is shown that formula 2 has chain scission occur after it is embedded in soil for 4 months (peak of chain scission is around ~1560 cm^{-1}) [20].

The absorption frequencies of functional group were also shown in Appendix-A.

4.4. Water Uptake

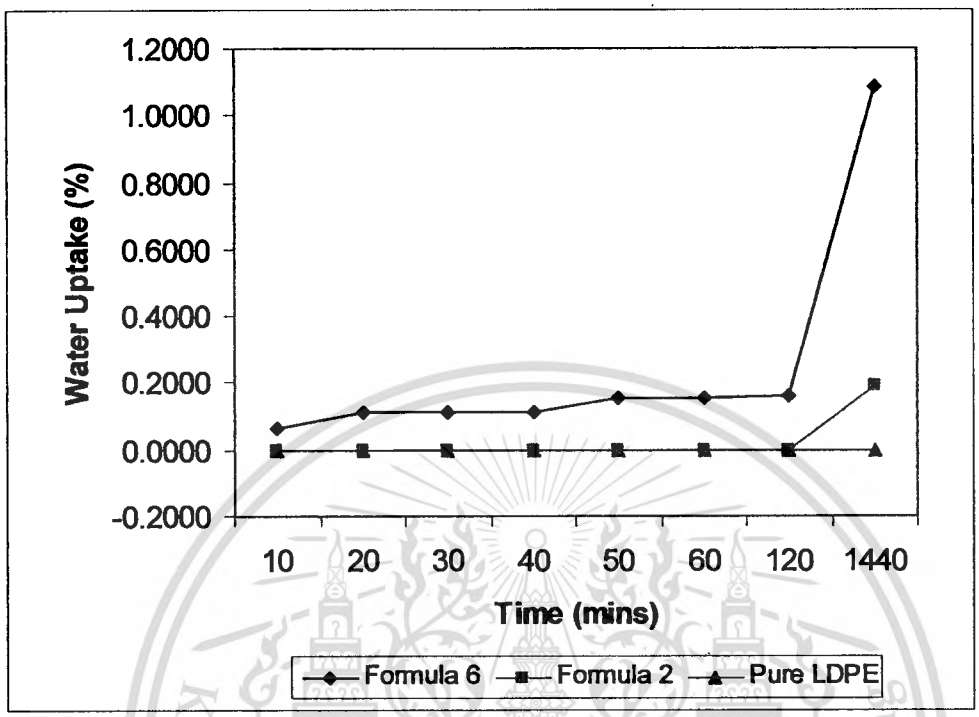


Figure 4.13: Relationship of water uptake (%) versus immersion time (min) of formula 2, formula 6 and pure LDPE.

Figure 4.13 is shown the water uptake for samples of formula 2, 6 and LDPE during 24 hours. A film sample from formula 2 were changed a little at 24 hour as formula 6 was the highest changed in volume (water uptake of formula 6 = 1.0881%).

In case of LDPE film, it has no water uptake after film is immersed under water for 24 hours. However, film from formula 6 has 1.08% of water uptake. This was due to mucilage powder in formula 6 was higher than in formula 2. The result confirmed that mucilage powder could be act as water storage.

The results of weight change in 24 hour and water uptake (%) at 24 hour of LDPE, formula 2 and formula 6 were also shown in Appendix-B.

4.5. Measurement of Tensile and Elongation at break

The mechanical properties of LDPE and LDPE:PEG:mucilage are shown in figure 4.14.

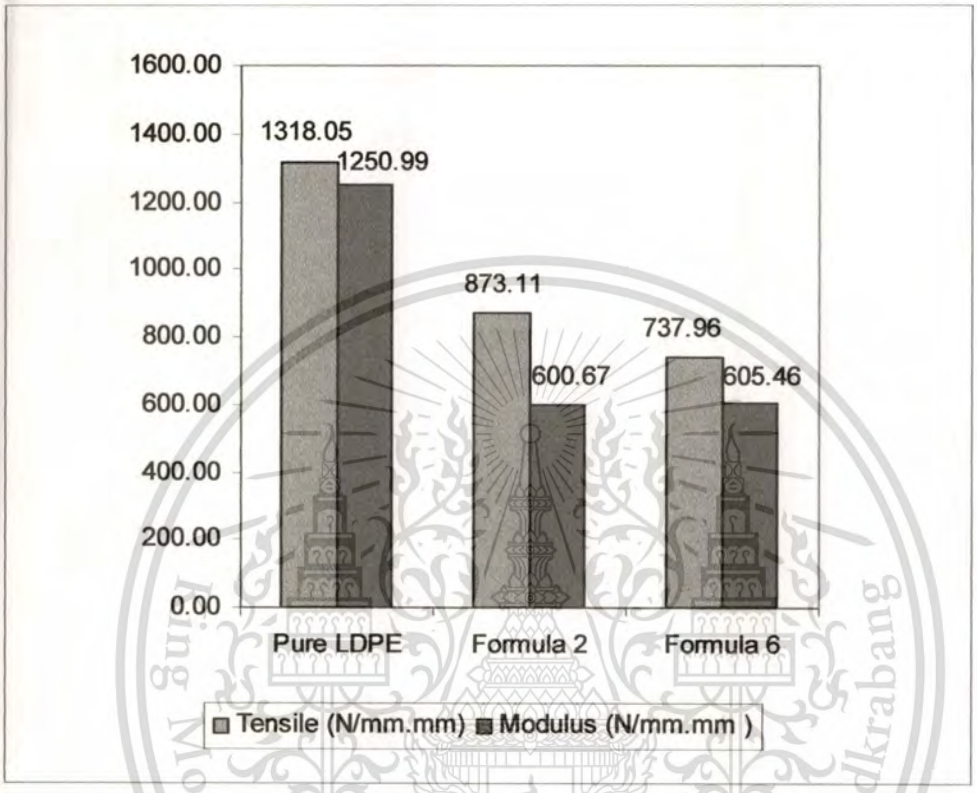


Figure 4.14 Tensile strength and modulus in the machine direction (MD) of pure LDPE, formula 2 and formula 6

Tensile strength and modulus were decreased as mucilage content was increased.(Figure 4.14)

As illustrated in the Figure 4.14, tensile strength was decreased from 1318 N/mm^2 to 837 N/mm^2 as 3.33% of PEG was present in formula 2. However, when 5% of PEG (formula 6) was added into LDPE, the tensile strength slightly decreased from 873 N/mm^2 to 737 N/mm^2. The reduction of tensile strength with addition of the PEG has appeared less efficient when it is compared with the other material properties.

The more flexibility of PEG compared to LDPE matrix, leading to more free volume of matrix. Tensile strength generally decreased after impact modifier was

added. The liquid PEG grade, particularly PEG 400, was suitable as plasticizer. When modified mucilage powder with PEG 400, the film was decreased in tensile strength.

The modulus of film was also decreased to 600 N/mm^2 in formulas, 2 and 6. It could be explained that flexibility of the films was improved by the role of PEG:mucilage (1:1).

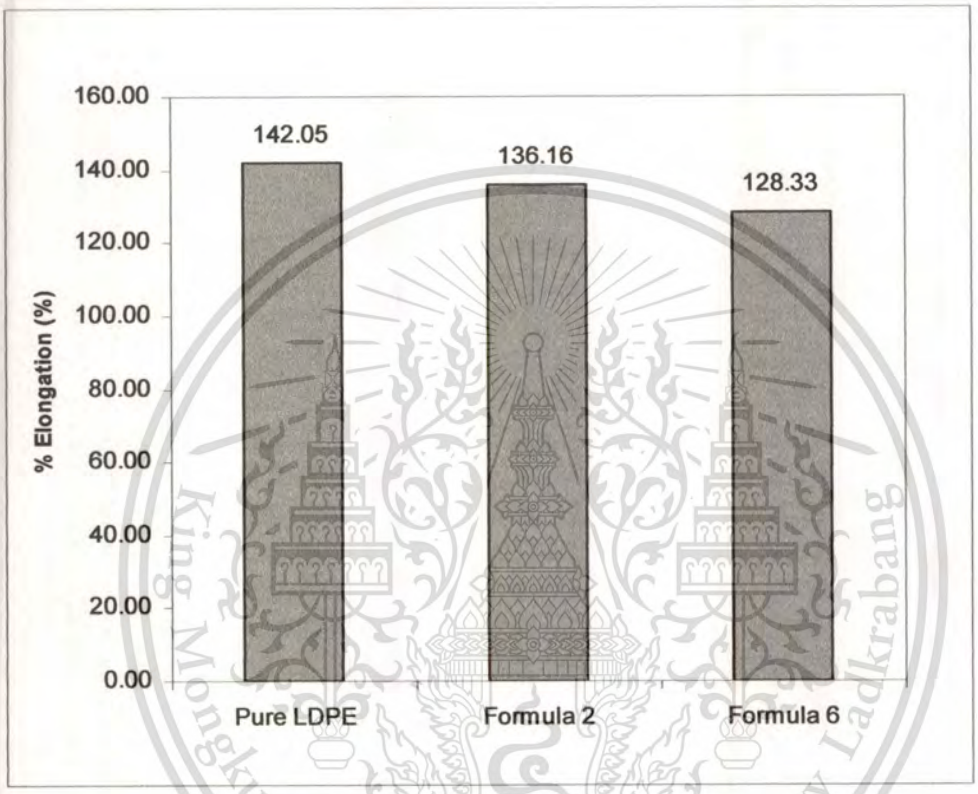


Figure 4.15 %Elongation in the machine direction (MD) of pure LDPE, formula 2 and formula 6.

As generally observed, the addition of mucilage reduced the elongation of the composites, as shown in Figure 4.15. The effect was more pronounced at higher mucilage content, formula 6 (PEG15:mucilage15). It was considered that the decrease in composite ductility depending on the structural rigidity of mucilage that was interfering during LDPE matrix extension.

PEG increased ductility of the composites while the mucilage decreased.

Results and the comparison of tensile strength, modulus and %elongation in both machine direction (MD) and transverse direction (TD) of pure LDPE, formula 2

(10PEG-10mucilage) and formula 6(15PEG-15mucilage) were also shown in Appendix-C.

4.6. Soil burial Test

In the experiment, we put film of formula 2 (10PEG:10mucilage) under soil for 4 months.

Figure 4.16 The film samples formula 2 before embedded in soil

Figure 4.17 The film samples formula 2 were embedded in soil for 1 month

Figure 4.18 The film samples formula 2 were embedded in soil for 4 month.

The photograph from digital camera was shown in Figure 4.16-4.18. Surface deterioration of the sample film could not be seen clearly from the photograph because of the low sensitivity of digital camera

However, the photograph was implied that increasing the time for soil burial test leading to decrease in degree of transparency of film sample. The more opaque of film probably may come from soil adhesion on the surface. The more unclear of film indicated that the film might be surface deterioration.

Photograph from microscope before and after embedded were also shown in Appendix-D.

4.6.1 Weight lost

Film formula 2 was cut into 4.5x7.5 cm. pieces then embedded in soil. Every month the film was weighed to determine the weight loss (Table 4.3)

Table 4.3 Weight of film before and after soil burial test

Film number	Weight of film (g.)			
	Before buried	Buried 1 month	Buried 2 months	Buried 4 months
1	0.3468	0.3313	-	-
2	0.3178	0.3123	-	-
3	0.3237	0.3132	0.2823	-
4	0.3303	0.3232	-	0.2780

Table 4.4 Percent weight lost of film before and after soil burial test

Film number	Weight lost (%)
1 (not buried)	0.00
2 (buried 1 month)	1.73
3 (buried 2 months)	12.79
4 (buried 4 months)	15.83

Table 4.4 could be explained about the effect of microorganism with LDPE/mucilage film. After 4 months in soil LDPE/mucilage film had weight lost of 15.83%. When time is increased in soil burial, the tendency of percent weight lost was increased. Because of mucilage powder in the LDPE was source of food for microorganism in soil. The microorganism could attach surface of film and show the evidence of LDPE surface opaque.

Chapter 5

Conclusion and Recommendation

5.1 Conclusion

The properties of films prepare from low density polyethylene blends with mucilage gel of *Ocimum canum Sims* and polyethylene glycol (M.W.400) can be concluded as follow:

1. Among 6 formulae, only formula 2 (mucilage = 3.33%) and formula 6 (mucilage = 5.0%) could be prepared by blown film process. This means that the optimum mucilage powder should be 5.00%.

2. MFI has no effect to the process because formula 2 and 6 which could be prepared blown film have different in MFI and formula 3(MFI = 4.26 g / 10mins) which has closely MFI of formula 2(MFI = 4.29 g/10mins) could not prepare blown film.

3. When consider the ratio of plasticizer (polyethylene glycol) with mucilage powder found that, appropriate ratio for preparation of blown film is 1:1 (formula 2 and formula 6).

4. Comparison of formula 4, 5, and 6 which containing the same amount of PEG but different amount of mucilage powder, it should be noted that the more content of mucilage powder (formula 4) should indicate the lowest MFI.

5. The thermogram of mucilage powder is shown the highest weight loss at 232°C. For the low density polyethylene, the highest weigh lost was at 478°C. The other mixture thermograms are shown of low density polyethylene with mucilage powder and polyethylene glycol the two highest weight lost around 300°C and 470°C. At around 470°C is the position of weight lost of low density polyethylene, The higher the amount of mucilage powder the lower the decomposition temperature of LDPE.

6. Formula 2 is shown chain scission after it is embedded in soil for 4 months (peak of chain scission is around ~1560 cm^{-1})

7, In case of LDPE film, it has no water uptake after film is immersed under water for 24 hours. However, film from formula 6 had 1.08% of water uptake. This was due to mucilage powder in formula 6 was higher than in formula 2. The result confirmed that mucilage powder could be act as water storage.

8. Decreasing of tensile and modulus depend on increasing of mucilage powder and polyethylene glycol content. Tensile and modulus value could arrange from high to low as follow: pure LDPE > LDPE formula 2 (10PEG:10mucilage) > LDPE formula 6 (15PEG:15mucilage) respectively.

5.2 Recommendation

1. Use compatibilizer that has melting temperature around T_m of low density polyethylene for make LDPE/mucilage compound well dispersed and distributed.

2. Make soil burial test with every formula for compare the ability of film degradation.

3. Make soil burial test at least 12 months.

4. Should make water uptake test more than 24 hours.

5. Add pigment in mixing process if the appearance of film is not beautiful.

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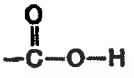
Appendix-A

FT-IR

Table A-1: The absorption frequency of functional group [20]

Type of functional group	**Formula**	**Absorption frequency (cm^{-1})**
Free hydroxyl	-OH	3638
Intermolecular hydrogen bond	-OH	3400-3200
In-plane bending hydroxyl		1420-1200
Methylene	$-CH_2$	2925±10 Asym. Streching 2850±10 Sym. Streching 1465±10 Asym. Bending
Ether	$-CH_2-O-CH_2-$	1150-1060 Asym. Streching
Double bonded carbon	$-CH=CH_2$	1655-1650
Carbonyl of carboxylic	C=O	1710±10
Polyene	$-(C=C)_n-$	1650-1580

Appendix-B

Water Uptake

Table B-1: The results of weight change in 24 hour and water uptake (%) at 24 hour of pure LDPE

Sample No.	Weight (g)									Water Uptake @24 hr (%)
	Time (minutes)									
	0	10	20	30	40	50	60	120	24 hr	
1	0.0766	0.0766	0.0766	0.0766	0.0766	0.0766	0.0766	0.0766	0.0766	0
2.	0.0753	0.0753	0.0753	0.0753	0.0753	0.0753	0.0753	0.0753	0.0753	0
3.	0.077	0.077	0.077	0.077	0.077	0.077	0.077	0.077	0.077	0
4.	0.0652	0.0652	0.0652	0.0652	0.0652	0.0652	0.0652	0.0652	0.0652	0
5.	0.0784	0.0784	0.0784	0.0784	0.0784	0.0784	0.0784	0.0784	0.0784	0
									Mean	0

Table B-2: The results of weight change in 24 hour and water uptake (%) at 24 hour of formula 2.

Sample No.	Weight (g)									Water Uptake @24 hr (%)
	Time (minutes)									
	0	10	20	30	40	50	60	120	24 hr	
1	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0
2.	0.0762	0.0762	0.0762	0.0762	0.0762	0.0762	0.0762	0.0762	0.0764	0.2625
3.	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0.0718	0.072	0.2786
4.	0.0776	0.0776	0.0776	0.0776	0.0776	0.0776	0.0776	0.0776	0.0777	0.1290
5.	0.0732	0.0732	0.0732	0.0732	0.0732	0.0732	0.0732	0.0732	0.0734	0.2733
									Mean	0.1886

Table B-3 The results of weight change in 24 hour and water uptake (%) at 24 hour of formula 6.

Sample No.	Weight (g)									Water Uptake @24 hr (%)
	Time (minutes)									
	0	10	20	30	40	50	60	120	24 hr	
1	0.2062	0.2065	0.2065	0.2065	0.2065	0.2067	0.2067	0.2067	0.2084	1.0669
2.	0.2266	0.2267	0.2267	0.2267	0.2267	0.2268	0.2268	0.2268	0.2289	1.0150
3.	0.1965	0.1966	0.1966	0.1966	0.1966	0.1966	0.1966	0.1967	0.1991	1.3232
4.	0.2322	0.2323	0.2327	0.2327	0.2327	0.2328	0.2328	0.2328	0.2348	1.1197
5.	0.2512	0.2513	0.2515	0.2515	0.2515	0.2515	0.2515	0.2515	0.2535	0.9156
									Mean	1.0881

Table B-4: The results of water uptake (%) of formula 6 in 24 hours.

Sample No.	Weight (g)								
	Time (minutes)								
	0	10	20	30	40	50	60	120	24 hr
1	0.1455	0.1455	0.1455	0.1455	0.2425	0.2425	0.2425	1.0669	0.1455
2.	0.0441	0.0441	0.0441	0.0441	0.0883	0.0883	0.0883	1.0150	0.0441
3.	0.0509	0.0509	0.0509	0.0509	0.0509	0.0509	0.1018	1.3232	0.0509
4.	0.0431	0.2153	0.2153	0.2153	0.2584	0.2584	0.2584	1.1197	0.0431
5.	0.0398	0.1194	0.1194	0.1194	0.1194	0.1194	0.1194	0.9156	0.0398
Mean	0.0647	0.1151	0.1151	0.1151	0.1519	0.1519	0.1621	1.0881	0.1151

Appendix-C

Mechanical Properties

Table C-1 The results of tensile test of pure LDPE in the machine direction (MD).

Sample No.	Max. Load (N)	Tensile (N/mm^2)	% Elongation	Modulus (N/mm^2)	Area (mm^2)
MD 1.	6.0560	1296.29	148.48	873.04	0.0047
MD 2.	5.6050	1387.34	122.72	1130.49	0.0040
MD 3.*	5.0130	1196.13	43.80	2730.90	0.0042
MD 4.*	6.2360	1368.74	209.08	654.65	0.0046
MD 5.	5.8210	1341.74	154.96	865.86	0.0043
Mean	5.7462	1318.0496	142.0533	1250.99	0.0044

Table C-2 The results of tensile test of pure LDPE in the transverse direction (TD).

Sample No.	Max. Load (N)	Tensile (N/mm^2)	% Elongation	Modulus (N/mm^2)	Area (mm^2)
TD 1.	4.7070	1097.71	56.16	1954.62	0.0043
TD 2.	4.6560	1163.42	81.56	1426.46	0.0040
TD 3.	4.5910	900.20	54.28	1658.43	0.0051
TD 4.*	5.3050	1343.65	33.22	4045.18	0.0039
CD 5.	5.5080	993.51	49.68	1999.81	0.0055
Mean	5.0278	1083.7660	48.3340	2414.5105	0.0046

* Error value didn't use it in calculation and discussion.

Table C-3 The results of tensile test of formula 2 (10PEG-10mucilage in the machine direction (MD).

Sample No.	Max. Load (N)	Tensile (N/mm^2)	% Elongation	Modulus (N/mm^2)	Area (mm^2)
MD 1.	7.5120	920.1890	147.9600	621.92	0.0082
MD 2.*	6.9260	686.4657	97.0400	707.40	0.0101
MD 3.*	7.3900	739.3734	198.6400	372.22	0.0100
MD 4.	7.1940	869.5252	124.3600	699.20	0.0083
MD 5.*	8.6800	829.6296	172.5200	480.89	0.0105
Mean	7.35	894.86	136.16	660.56	0.0094

Table C-4 The results of tensile test of formula 2 (10PEG-10mucilage) in the transverse direction (TD).

Sample No.	Max. Load (N)	Tensile (N/mm^2)	% Elongation	Modulus (N/mm^2)	Area (mm^2)
TD 1.	4.7600	660.6523	84.2400	784.25	0.0072
TD 2.	5.4880	685.1094	110.9200	617.66	0.0080
TD 3.	3.4070	699.9918	10.6800	6554.23	0.0049
TD 4.	3.2040	734.3234	10.2680	7151.57	0.0044
TD 5.	4.5260	670.8365	74.7200	897.80	0.0067
Mean	4.2770	690.1827	58.1656	3201.10	0.0062

* Error value didn't use it in calculation and discussion.

Table C-5 The results of tensile test of formula 6 (15PEG-15mucilage) in the machine direction (MD).

Sample No.	Max. Load (N)	Tensile (N/mm^2)	% Elongation	Modulus (N/mm^2)	Area (mm^2)
MD 1.	5.9900	739.5308	167.9200	440.41	0.0081
MD 2.*	6.1310	629.8541	98.6400	638.54	0.0097
MD 3.	7.1780	765.8527	98.1600	780.21	0.0094
MD 4.	5.0960	708.4863	118.9200	595.77	0.0072
MD 5.*	5.2020	546.4349	43.0000	658.36	0.0095
Mean	6.0880	737.9566	128.3333	605.4608	0.0088

Table C-6 The results of tensile test of formula 6 (15PEG-15mucilage) in the transverse direction (TD).

Sample No.	Max. Load (N)	Tensile (N/mm^2)	% Elongation	Modulus (N/mm^2)	Area (mm^2)
TD 1.*	5.7200	705.0291	237.1600	297.28	0.0081
TD 2.*	4.0940	489.3187	30.4840	1605.17	0.0084
TD 3.	6.1520	628.2918	74.8400	839.51	0.0098
TD 4.*	4.3410	514.0502	38.7360	1327.06	0.0084
TD 5.	6.8900	597.3643	89.4400	667.89	0.0115
Mean	6.5210	612.8280	82.1400	753.7036	0.0093

* Error value didn't use it in calculation and discussion.

Figure C-1 Tensile strength and modulus in the transverse direction (TD) of pure LDPE, formula 2 (10PEG-m10ucilage) and formula 6(15PEG-15mucilage)

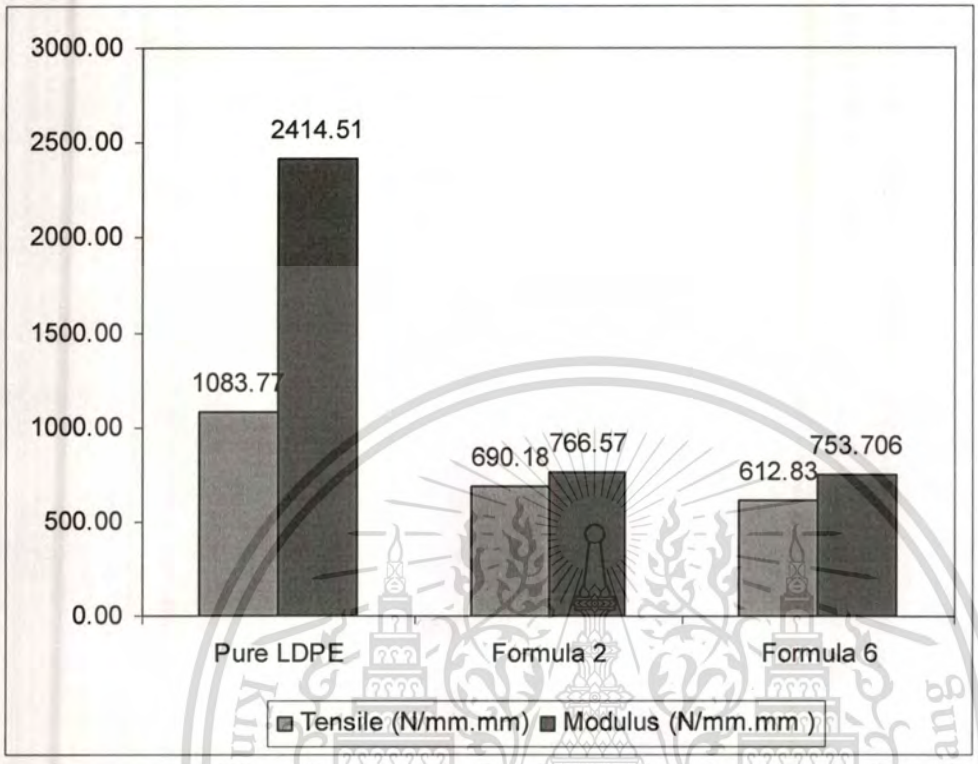


Figure C-2 %Elongation in the transverse direction (TD) of pure LDPE, formula 2 (10PEG-10mucilage) and formula 6(15PEG-15mucilage)

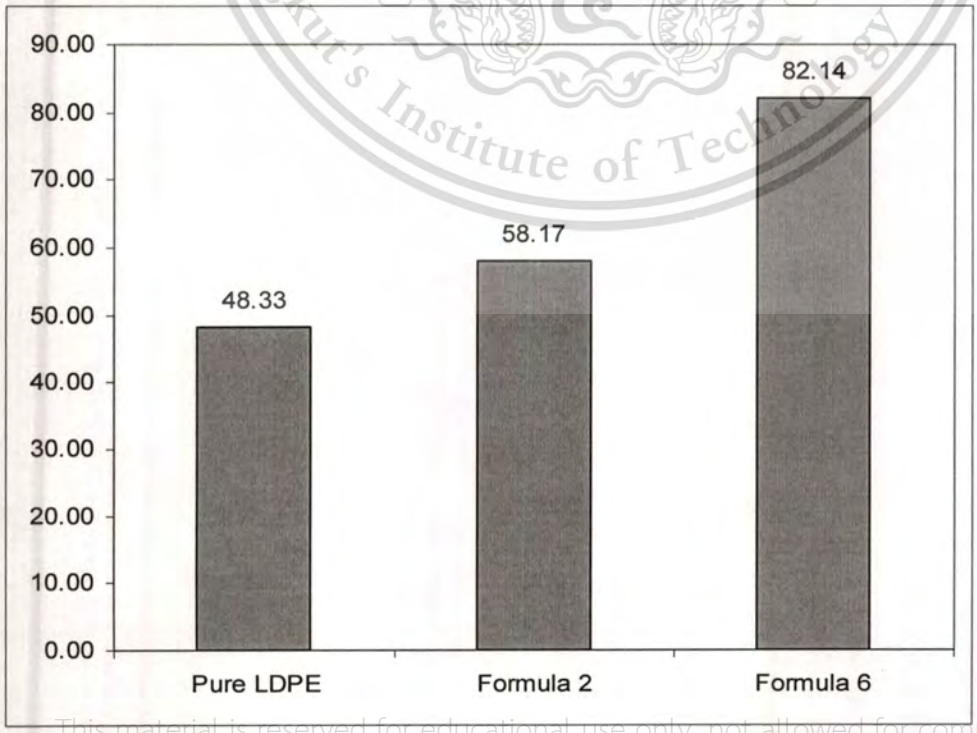


Figure C-3 The comparison of tensile strength in machine direction (MD) and transverse direction (TD) of pure LDPE, formula 2 (10PEG-10mucilage) and formula 6(15PEG-15mucilage)

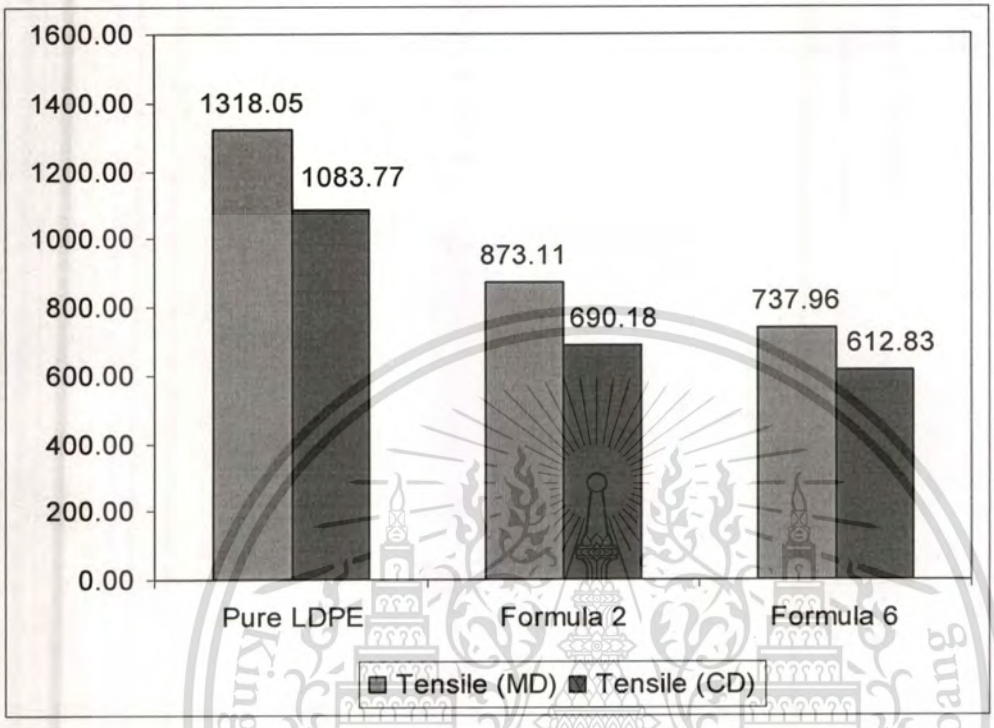


Figure C-4 The comparison of modulus in machine direction (MD) and transverse direction (TD) of pure LDPE, formula 2 (10PEG-10mucilage) and formula 6(15PEG-15mucilage)

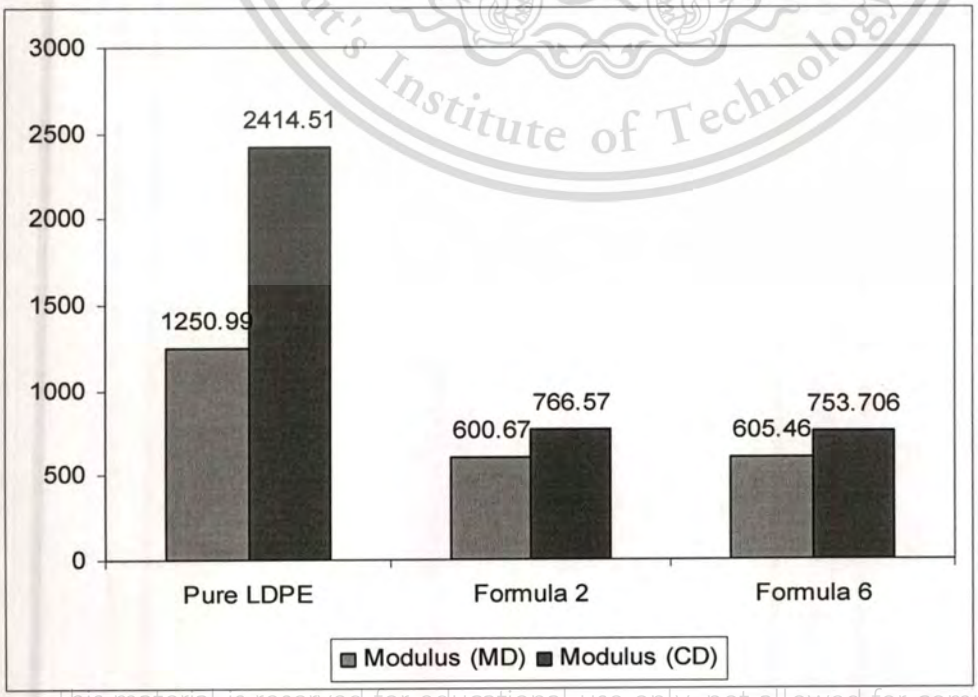
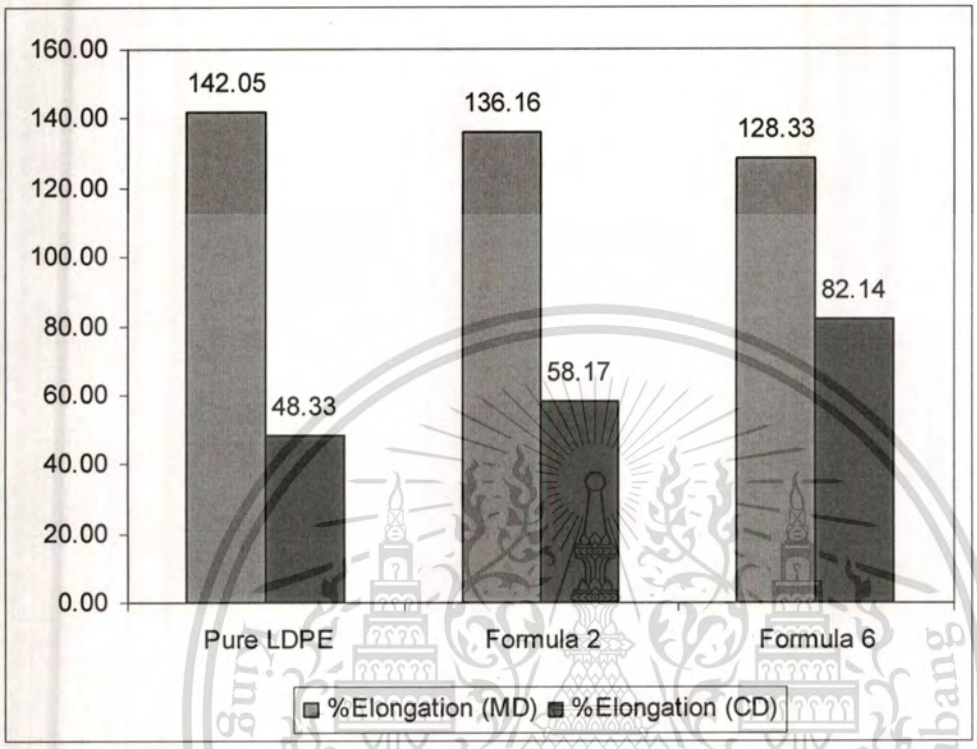


Figure C-5 The comparison of elongation in machine direction (MD) and transverse direction (TD) of pure LDPE, formula 2 (10PEG-10mucilage) and formula 6(15PEG-15mucilage)



Appendix-D

Microscope

Appendix-C

Mechanical Properties

Figure D-1 Microscope showing the surface of film samples formula 2 (PEG10:mucilage10) before embedded under the soil (×20 objective lens and ×10 eyepiece lens).

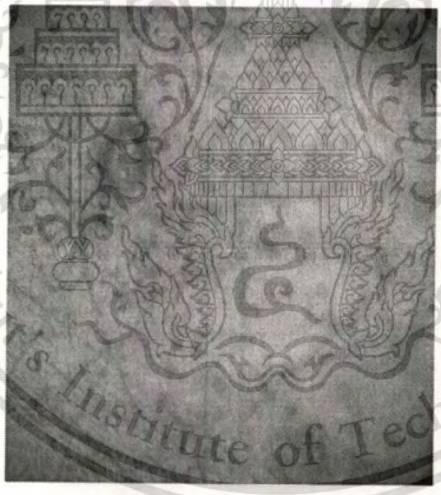


Figure D-2 Microscope showing the surface of film samples formula 2 (PEG10:mucilage10) after embedded under the soil for 1 month (×20 objective lens and ×10 eyepiece lens).

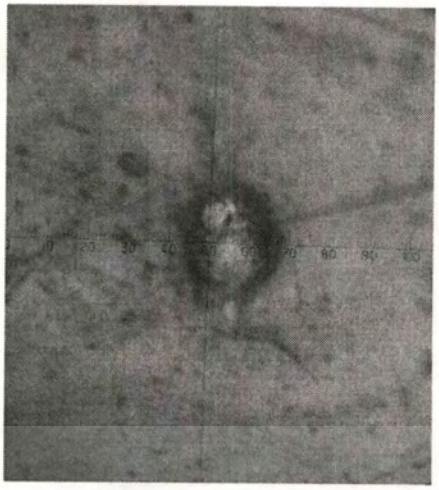


Figure D-3 Microscope showing the surface of film samples formula 2 (PEG10:mucilage10) after embedded under the soil for 2 month (×20 objective lens and ×10 eyepiece lens).

Figure D-4 Microscope showing the surface of film samples formula 2 (PEG10:mucilage10) after embedded under the soil for 4 months (×20 objective lens and ×10 eyepiece lens).