UNIVERSITI PUTRA MALAYSIA

CHARACTERIZATION AND STORAGE STUDIES OF SOLUBLE PAPAYA PUREE PECTIN-BASED FILMS ON AGAR-AGAR POWDER

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ABSTRACT

This study investigates the potential use of soluble papaya puree pectin-based films on stored agar-agar powder. Two objectives of the study were to characterize the soluble papaya puree pectin-based films properties, including water solubility, tensile strength, seal strength, water vapour permeability, surface roughness and optical, and to evaluate the effect of the films on the quality changes in stored agar-agar powder. The soluble papaya puree pectin-based film was made using a formulation ratio of 1:0.5:1 (pectin:glycerol:papaya puree). The formulation for soluble pectin film (control) is 1:0.5 (pectin:glycerol). Water solubility test, texture analyzer, water vapour permeation test, atomic force microscopy and colorimeter were used to characterize the films. The agar-agar powders were stored for 39 days at ambient (28 °C) and chill (4 °C) temperatures. At selected storage time, moisture content, true density and colour of agar-agar powder were evaluated using moisture analyzer, gas pycnometer and colorimeter, respectively. The results revealed that soluble pectin film has higher solubility percentage than soluble papaya puree pectin-based film where the solubility percentage of soluble pectin film and papaya puree pectin-based film were 93.7 ± 5.8% and 84.5 ± 5.2%, respectively. The tensile properties of the soluble papaya puree pectin-based film were 4.8 ± 0.1 MPa for tensile strength (TS), 54.9 ± 3.7% for elongation at break (EAB), 0.17 ± 0.0 for Young Modulus (YM) and 1.77 ± 0.02 MJ/m^3 for toughness. Meanwhile, the soluble pectin film showed values of TS, EAB, YM and toughness of 4.3 ± 0.1 MPa, 42.1 ± 1.2%, 0.19 ± 0.0 and 1.1 ± 0.0 MJ/m^3, respectively, which were slightly lower that the soluble papaya puree pectin-based film. The soluble papaya puree pectin-based film also had better seal strength compared to the soluble pectin film, which papaya puree pectin-based film had seal strength of 0.5 ± 0.1 N/mm while pectin film was 0.4 ± 0.0 N/mm. Moreover, the water vapour permeability (WVP) of soluble papaya puree pectin-based film was better than soluble pectin film, where the WVP value of papaya puree pectin-based film and pectin film were 115.5 E-08 g /m. day Pa ± 9.9 to 116.3 E-08 g /m. day Pa ± 33.2, respectively. Also, the optical properties of the soluble papaya puree pectin-based films showed that it is less transparent compared to soluble pectin film. The AFM test revealed that soluble papaya puree pectin film has rougher surface that soluble pectin film. For the studies of quality changes on the agar-agar powder at selected storage temperature, the moisture content of agar-agar powder packed with the soluble papaya puree pectin-based film was lower than the one packed with soluble pectin film at both storage temperatures. Meanwhile, the true density of the agar-agar powder showed decreasing trend on both storage temperatures and types of soluble film. The colour of agar-agar powder also showed degradation on both storage temperature. The decrement of total colour difference (ΔE) on both storage temperatures and types of film indicates that the agar-agar powder loses its yellowish colour intensity during the 39 days storage time. As a conclusion, both objectives have been achieved where the characteristic of pectin film has been enhanced by the addition of papaya puree except the water solubility, surface roughness and optical properties of the film. Also, the quality changes of agar-agar powder when stored in papaya puree pectin-based film shows less changes compare to when stored in pectin film, except the color of the agar-agar powder which is whiter when stored in papaya puree pectin-based film.

ABSTRAK

Kajian ini menyelidiki potensi penggunaan filem berasaskan pektin betik larut pada serbuk agar-agar yang disimpan. Dua objektif kajian adalah untuk mencirikan sifat filem berdasarkan pektin betik yang larut, termasuk kelarutan dalam air, kekuatan tegangan, kekuatan meterai, kebolehtelapan wap air, kekasaran permukaan dan optik, dan untuk menilai kesan filem terhadap perubahan kualiti dalam serbuk agar-agar yang disimpan. Filem berdasarkan pektin betik larut peptik dibuat menggunakan nisbah formulasi 1: 0.5: 1 (pektin: gliserol: puri betik). Rumusan untuk filem pektin larut (kawalan) adalah 1: 0.5 (pektin: gliserol). Uji kelarutan air, penganalisis tekstur, uji permeasi uap air, mikroskopi kekuatan atom dan kolorimeter digunakan untuk mencirikan filem. Serbuk agar-agar disimpan selama 39 hari pada suhu sekitar (28 ° C) dan suhu sejuk (4 ° C). Pada waktu penyimpanan yang dipilih, kandungan kelembapan, kepadatan sejati dan warna serbuk agar-agar dinilai masing-masing menggunakan penganalisis kelembapan, pycnometer gas dan colorimeter. Hasil kajian menunjukkan bahawa filem pektin larut mempunyai peratusan kelarutan yang lebih tinggi daripada filem berasaskan pektin betik larut di mana peratusan kelarutan filem pektin larut dan filem berdasarkan pektin betik puri adalah 93.7 ± 5.8% dan 84.5 ± 5.2%. Sifat tegangan filem berasaskan pektin betik yang larut ialah 4.8 ± 0.1 MPa untuk kekuatan tegangan (TS), 54.9 ± 3.7% untuk pemanjangan semasa rehat (EAB), 0.17 ± 0.0 untuk Young's Modulus (YM) dan 1.77 ± 0.02 MJ / m^3 untuk ketangguhan. Sementara itu, filem pektin larut menunjukkan nilai TS, EAB, YM dan daya tahan 4.3 ± 0.1 MPa, 42.1 ± 1.2%, 0.19 ± 0.0 dan 1.1 ± 0.0 MJ / m^3, masing-masing, yang sedikit lebih rendah daripada pektin pekat betik larut filem berdasarkan. Filem betik larut pektin juga mempunyai kekuatan meterai yang lebih baik berbanding dengan filem pektin larut, dimana filem pektin pektik betik mempunyai kekuatan meterai 0,5 ± 0,1 N / mm sementara filem pektin 0,4 ± 0,0 N / mm. Lebih-lebih lagi, kebolehtelapan wap air (WVP) filem berdasarkan pektin betik larut peptik lebih baik daripada filem pektin larut, di mana nilai WVP filem berasaskan pektin betik puri dan filem pektin adalah 115.5 E-08 g / m. hari Pa ± 9,9 hingga 116,3 E-08 g / m. hari Pa ± 33.2, masing-masing. Selain itu, sifat optik dari filem berasaskan pektin betik larut menunjukkan bahawa ia kurang telus berbanding dengan filem pektin larut. Ujian AFM menunjukkan bahawa filem pektin betik larut mempunyai permukaan yang lebih kasar daripada filem pektin larut. Untuk kajian perubahan kualiti pada serbuk agar-agar pada suhu penyimpanan yang dipilih, kandungan kelembapan serbuk agar-agar yang dibungkus dengan filem berasaskan pektin betik larut lebih rendah daripada yang dibungkus dengan filem pektin larut pada kedua suhu penyimpanan. Sementara itu, ketumpatan sebenar serbuk agar-agar menunjukkan kecenderungan penurunan pada suhu penyimpanan dan jenis filem larut. Warna serbuk agar-agar juga menunjukkan penurunan pada kedua-dua suhu penyimpanan. Penurunan perbezaan warna total (ΔE) pada suhu penyimpanan dan jenis filem menunjukkan bahawa serbuk agar-agar kehilangan intensiti warna kekuningannya selama 39 hari waktu penyimpanan. Sebagai kesimpulan, kedua-dua objektif telah dicapai di mana ciri filem pektin telah ditingkatkan dengan penambahan puri betik kecuali kelarutan air, kekasaran permukaan dan sifat optik filem. Juga, perubahan kualiti serbuk agar-agar ketika disimpan dalam filem berasaskan pektin puri betik menunjukkan perubahan yang lebih sedikit dibandingkan dengan ketika disimpan dalam filem pektin, kecuali warna serbuk agar-agar yang lebih putih ketika disimpan dalam filem pektin betik larut peptik.

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CHAPTER 1

INTRODUCTION

1.1 BACKGROUND STUDY

The forecast reveals a number of distinct trends as we get closer to 2021, ranging from sustainability and environmentally friendly packaging to "smart packaging" and beyond. Sustainability and environmentally friendly packaging are two types of packaging that are expected to gain popularity among consumers in the coming years. Sustainable packaging is defined as the sourcing, development, and use of packaging solutions that are low-impact on the environment. Sustainable packaging is eco-friendly and does not contribute to the depletion of natural resources.

As the millennial and "Z" generations enter the workforce and become more consumers, sustainability, recycling, and environmental issues will continue to take center stage. Consumers are anticipated to demand more ecologically friendly packaging in the future, from zero-waste packaging reforms to reusable packaging programmes. The terms of "reduce, reuse, recycle" is only anticipated to become louder as the world becomes more aware of the environmental implications of packaging supplies. Due to the tremendous challenges linked with plastics and other waste products that have found their way into our oceans, businesses will be required to offer environmentally appropriate alternatives (*The Time Is Now for Edible Packaging*, n.d.)

In recent years, primary food packaging has largely consisted of passive materials that act as an inert barrier to the entry of oxygen and moisture into the food product. However, retailers, manufacturers, and eCommerce businesses are increasingly considering plastic alternatives. One European company, for example, has produced water-soluble and compostable shopping bags, clothing bags, laundry bags, and packaging film. Soluble packaging has steadily acquired producer and consumer interest as a means of minimizing plastic pollution over the years (Water-Soluble Packaging: A New Plastic Alternative - EcoBahn, n.d.). Soluble packaging can also be used to improve the appearance of food while reducing synthetic polymer use.

Soluble films, on the other hand, are food wrappers comprised of a thin layer of material that may be eaten or removed, and are one of the most recent advances in the transition away from synthetic polymer-based packaging. As a result, it can be used to package dissolving solution products. Film-forming material, plasticizer, and additives are among the three major components of soluble film.

Polysaccharides have been used for soluble coatings and films in most studies because they are inexpensive and have good film-forming properties. Pectin is one of the polysaccharides that contribute to tissue integrity and rigidity for the production of soluble films among film-forming materials. Pectin's thickening and emulsifying properties, which solidify into a gel, make it popular in industry. Pectin derived from citrus peel or apple pomace is used as a stabilizer, gelling agent, crystallization inhibitor, and encapsulating agent in the food manufacturing. This polysaccharide is primarily made up of galacturonic acid units and, due to its chemical and structural characteristics, can interact with a variety of molecules. The characteristic structure of the pectin backbone is a linear chain of alpha (1-4) linked D-galacturonic acid (Thakur et al., 1997).

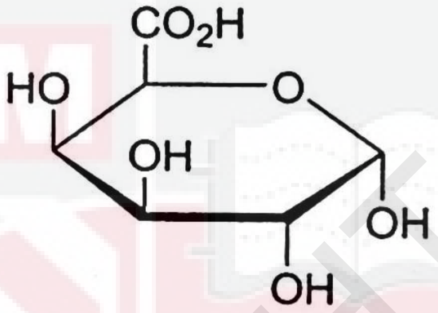
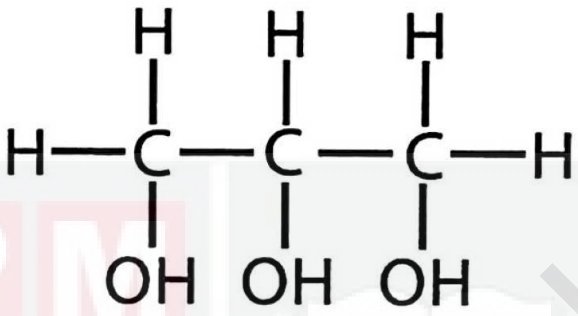


Figure 1.1: Molecular Structure of Pectin (Source: Thakur et al., 1997).

Rather than having high solubility, pectin has good hardness and adhesiveness and acts as an oxygen barrier. Therefore, pectin is a good candidate for effective biopolymer for producing soluble films because of its biodegradability, biocompatibility, and non-toxicity properties. Pectin is a good polymeric matrix for edible and soluble film for active food packaging, according to Valdés et al (2015). These hydrophilic compounds may provide effective barriers to oils and lipids, but they have poor moisture barrier properties.

Plasticizers in soluble coating formulations, such as glycerol, reduce the number of internal hydrogen bonds between polymer chains, allowing oxygen and water vapour to diffuse through the coating film (Valdés et al., 2015). For water-soluble polymers like pectin, this small polyol with three hydroxyl groups is the best plasticizer. When it comes to the mechanical properties of glassy composite systems, it has a higher plasticizing efficiency. Plasticizers work by reducing internal hydrogen bonds and breaking down the cohesion between film networks. Plasticizers were added to improve the film's workability and reduce brittleness (Sanyang et al., 2015). This plasticizer will be combined with pectin and fruit purees which are papaya puree, in the production of films to provide nutritional value, exploit fruit processing wastes, and reduce synthetic packaging production.



$$\mathrm{H-\overset{H}{\underset{OH}{C}}-\overset{H}{\underset{OH}{C}}-\overset{H}{\underset{OH}{C}}-H}$$

Figure 1.2: Structural formula of glycerol (Source: Langer, 1969)

Papaya fruit (Carica papaya) is a succulent fruit of a huge plant of Caricaceae's family. A fully ripe papaya's flesh becomes soft and succulent. It can also be eaten raw as a vegetable, sliced into thin strips, or processed into candy, pickles, and puree, among other things. Vitamins A, C, E, and K, as well as folate and fibre, are abundant in papayas. It is also fat-free, cholesterol-free, and sodium-free. A serving of fruit dish (1/2 papaya) contains only 70 calories on average (Milind et.al., 2011). Papaya flesh is a variety of colours ranging from pale yellow to deep red, with many shades in between.

Figure 1.3: Papaya fruit (Carica papaya) (Source: Kwok & Liang, 2019)

Pectin, found in papaya puree, can be used to create biodegradable, environmentally friendly edible films (Tharanathan, 2003). Sugars, pectin, and a few protein molecules are found in papaya control films (Sila et al., 2009). Papaya puree was used as a matrix for a soluble film because of its ability to reduce film rigidity and its appealing colour and flavour. The deep yellow or orange flesh of papaya has a slightly sweet taste, which improves sensory evaluation. Therefore, despite producing only a fraction of the world's output Malaysia's achievement of fourth place in world exports for exporting papaya fruit was surprising. Malaysia was the world's second-largest exporter in 2005, exporting 42,008 mt (Kwok & Liang, 2019). The increased papaya production will result in a significant amount of waste in Malaysia and worldwide.

1.2 PROBLEM STATEMENT

A major amount of contemporary food packaging materials is made of non-biodegradable synthetic plastics produced from non-renewable fossil resources. Production, usage, disposal, and accumulation have all been seen as severe hazards to our society and environment. To safeguard the environment and decrease the cost of combating plastic pollution, several countries and districts are gradually eliminating the use of single-use plastics in food packaging.

When it comes to waste disposal, the use of plastic packaging is always a consideration. How long can it be degraded, the environmental impact of the degradation process; and the most effective way to reduce plastic waste, environmental issues should always be a concern. To help alleviate these problems, biodegradable and renewable packaging, such as pectin, is being used.

Food waste has also become one of the primary sources of municipal waste (MSW) because of the high production of tropical fruits, posing increasing environmental challenges. Food is lost or wasted for various reasons, including bad weather, processing issues, overproduction, and volatile markets, resulting in food waste before it reaches the supermarket. In addition, fruit overproduction will result in more waste generation because it has a shorter shelf life.

This is because overripe fruits will not be consumed or even sold due to their soft shape and bruises, which make them unappealing and unpleasant to eat. In addition, these uneaten fruits cause unnecessary environmental damage by wasting valuable resources such as water and farmland. Papaya fruits were chosen for the project due to high post-harvest losses in the past. As a result, this project contributes to the reduction of environmental issues associated with fruit waste.

1.3 OBJECTIVES

The research objectives for this project are:

1. To compare the characteristics of papaya puree-based films made with a formulation ratio of 1: 0.5: 1 (pectin: glycerol: papaya puree) with pectin-based films (control film without papaya puree), including water solubility, tensile properties, seal strength, water vapour permeability, surface roughness. and optical properties.

2. To evaluate the effect of the films on the quality changes, including moisture content, true density and optical properties on stored agar-agar powder.

1.4 SCOPE OF STUDY

The research offers an opportunity to develop the papaya puree pectin-based films and compare them with the pectin-based film's characteristics. Also, it offers the chance to study the effect of the films on the quality changes in stored agar-agar powder by packing the agar-agar powder in the film at ambient (28 °C) and chill (4 °C) temperatures. The method for producing the pectin-based soluble film and the expected application of soluble film for packaging the agar-agar powder were demonstrated. The films were made into a small pack of dimensions 5 cm × 6 cm, where the agar-agar powders were inserted and sealed into a closed packet. Pectin film (as a control) and papaya puree pectin-based film were used as test films (pectin film added with papaya puree with the ratio of 1:1). The research work was carried out at Universiti Putra Malaysia's (UPM) Food Processing Quality Laboratory and Packaging and Shelf-Life Laboratory in Serdang, Selangor.

CHAPTER 2

LITERATURE REVIEW

2.1 SYNTHETIC POLYMERS PACKAGING

Human-made synthetic polymers are those that have been synthesized. The utility of thermoplastics, thermosets, elastomers, and synthetic fibers is divided into four categories. Thermoplastics are polymers that become moldable and malleable when heated and then solidify when cooled. Thermosets, on the other hand, harden once set and cannot be reshaped, making them popular in adhesives. A flexible polymer, also known as an elastomer or rubber, is a type of polymer that can be stretched or bent. Synthetic fibers are a type of polymer created by changing natural plant and animal fibers (7 Types of Plastic You Should Know – Waste4Change, n.d.).

The backbones of typical synthetic polymers like polythene and polystyrene are polyacrylates. They have carbon-carbon bonds, whereas hetero chain polymers, such as polyamides, polyesters, polyurethanes, polysulfides, and polycarbonates, have additional components incorporated into the backbone. Metals with non-covalent bonding make up the backbone of coordination polymers. Polyethylene terephthalate/polyester (PET), high-density polyethylene (HDPE), polyvinyl chloride (PVC), low-density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), and others are examples of modern plastics (Sherrington, 1992).

However, these plastics are causing serious environmental issues and are regarded as non-degradable. It may fragment into smaller pieces and migrate via river and ocean, forming accumulation zones along the way. These synthetic polymers can enter our environment through various routes, potentially clogging sewage systems and raising sewage maintenance costs. Furthermore, if synthetic polymers are not properly disposed of, they can endanger or kill animals if they are ingested or swallowed. It could also impact on soil fertility because it can accumulate in nature for decades without degrading due to the uncontrollable disposal method.

Even though it varies by type, any type of plastic exposed to extreme conditions, such as extreme heat, could leach hazardous materials. Three types of plastic are considered safer than the others: polyethylene terephthalate (PET), high-density polyethylene (HDPE), and polypropylene (PP). Although experts are currently working to develop the best method and strategy for recycling all of those types of plastic, the two types of plastic that are most commonly collected by recycling programmes are polyethylene terephthalate (PET) and high-density polyethylene (HDPE) (Grigore, 2017).

2.2 BIODEGRADABLE POLYMERS

Since the 1970s, degradable and/or biodegradable polymer materials have gotten much attention. The majority of food items are now packaged. As a result, biodegradable polymers are increasingly used in packaging in everyday life. Consumer demand, the spread of convenience packaging, the development of new uses for bioplastics, increasing economic viability as manufacturing ramps up and unit costs fall, and the construction of composting infrastructure have all profited from the rise in crude oil prices. These are the main factors that are propelling the biodegradable packaging market forward. Despite this, consumer demand for environmentally friendly, safer, and nontoxic products is expected to increase, and given the current favourable economic climate, biodegradable packaging products are expected to become more popular (Pawar & Purwar, 2013).

To reduce waste volume, biodegradable polymers are frequently used. In addition to biodegradability, biopolymers have other qualities such as air permeability, low temperature sealability, availability, and low cost. As shown in Figure 2.1, biopolymers such as starch, cellulose, chitosan, Poly (lactic acid) (PLA), Polyhydroxyalk anoates (PHA), poly-hydroxybutyrate (PHB), and others are utilized in packaging. Biopolymers are polymers that are typically biodegradable and non-toxic and are made from renewable natural resources. They can be made biologically (by microorganisms, plants, and animals) or chemically (from biological starting materials such as sugars, carbohydrates, natural fats and oils) (Ajay et al., 2019).

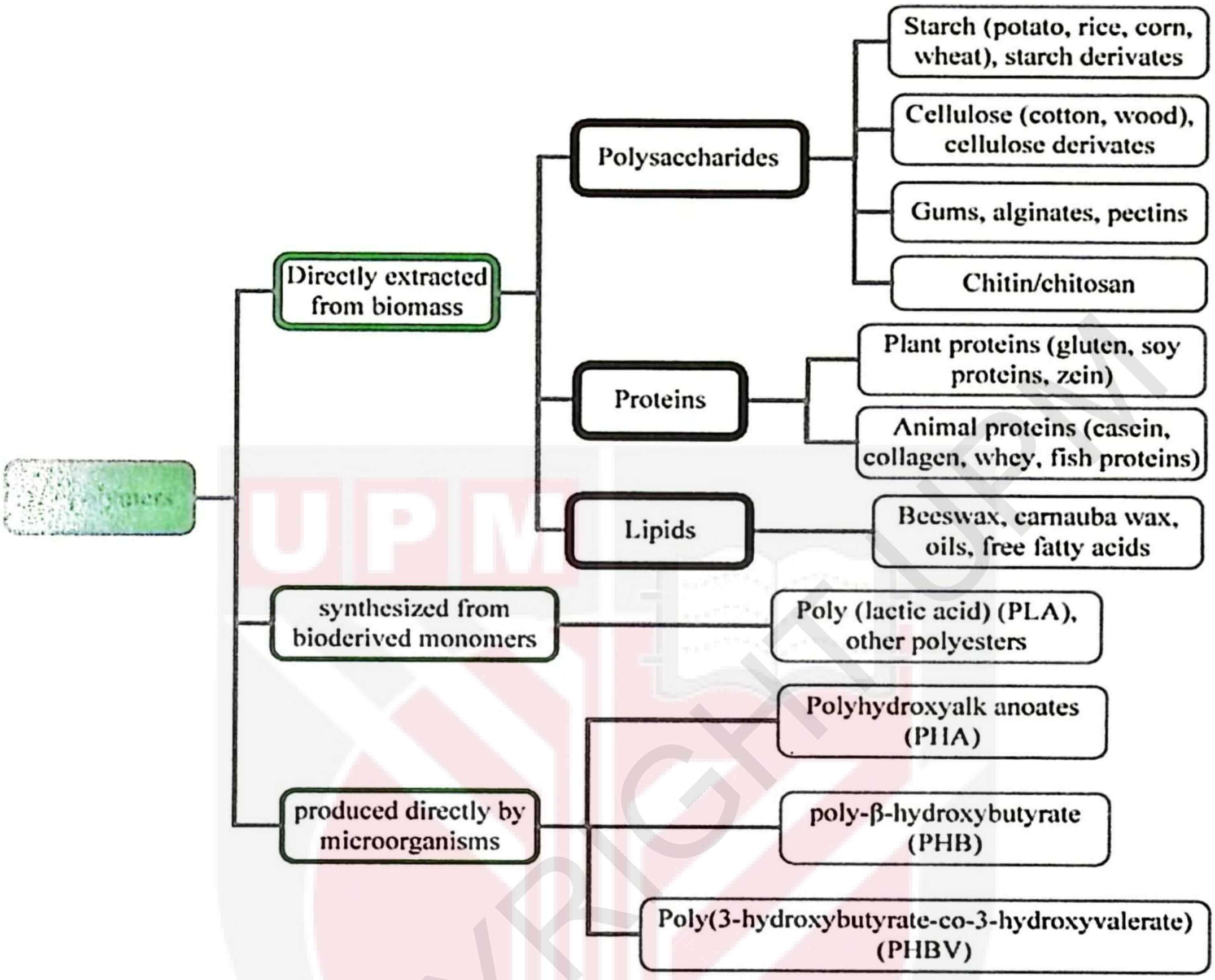


Figure 2.1: Schematic diagram of biopolymers (Source: Ajay et al., 2019)

Solubility refers to the ability of edible films to dissolve in water so that they can be properly digested when consumed or naturally decompose if released into the environment. Because soluble-film packaging melts in boiled water or in the mouth of the consumer, it is ideal for use in ready-to-eat food products Because high solubility properties of edible films cannot protect the product from humidity and water loss, the film's solubility should be tailored to the needs of the user (Arham et al., 2016).

The water-soluble film is a biodegradable plastic, which means that after dissolving in water, it leaves no toxic residue in the environment. Dissolution takes only a few minutes, and the film's components biodegrade into natural substances in less than three months. The water-soluble plastic film has a number of advantages in addition to its environmental benefits. The water-soluble plastic film helps to reduce waste by reducing the amount of packaging that needs to be discarded. In fact, unlike traditional packaging, we can get small unit doses of concentrated products by using a water-soluble packaging solution. As a result, the packaging size and waste generated are reduced. This reduction in packaging has a dual positive impact on the environment, as it allows us to reduce both plastic packaging and CO_2 emissions associated with bulky product transportation.

The biodegradable water-soluble film also has the advantage of allowing the user to hold a pre-dosed liquid or solid product in his hands. This unit dose simplifies and streamlines its application. With the water-soluble plastic film dose, there will be no waste. Finally, the packaging for some harsh products must be user-friendly. This is true for crop protection chemicals as well as chlorinated pool water treatment products. The user is protected by the water-soluble film, which reduces or eliminates any risk associated with handling or inhaling the product (*What Are the Benefits of Biodegradable Water Soluble Plastic Film ?*, n.d). Other functions and advantages of soluble film include shelf-life extension and safety enhancement. Food products with a higher protective function have a longer shelf life and are less likely to be contaminated by foreign matter.

2.3 PECTIN

In soluble films made from fruits and vegetables, pectin has long been the most common binding agent. In contrast to the food industry, where pectin is typically used as a stabilizer, thickening, and gelling agent, pectin could be used as a binding agent in the packaging industry to improve the film's properties (Otoni et al., 2017). Pectin is a high-molecular-weight white amorphous colloidal carbohydrate found in ripe fruits such as apples and currants. Its thickening and emulsifying capabilities, as well as its propensity to solidify to a gel, benefit fruit jellies, medications, and cosmetics. Pectin are plant-derived polysaccharide mixtures that contain pectinic acids as a major component, are water soluble, and can gel under the right conditions (BeMiller, 1986).

The carboxylic group sequences of esterified galacturonic acid molecules divide pectin into two groups. Pectin with a high DM (HM pectin) and galacturonic residues of 50% or higher are used in heat-resistant bakery jams, fruit preservatives, and juices. Due to hydrophobic interactions, HM pectin form gels in aqueous media under acidic conditions and a high sugar content. LM pectin with a low DM content is produced by de-esterification of HM pectin at a specific pH, temperature, and time. In LM pectin, the percentage of DM varies between 20% and 30%. They are used to make gels when divalent calcium is present and the pH is low. Low-sugar jams and jellies, dairy desserts, ice cream with fruit gels, syrup thickening agents for fruit and vegetable canning, and food coatings are all examples of food applications (Valdés et al., 2015).

As the gel strength increases, the tendency to gel, solubility, and pectin viscosity may all increase. Clumps may form as a result of the rapid hydration of agar-agar powdered pectin with water. Furthermore, when the pH is reduced, the polysaccharide chains will no longer repel against one another and will attach to form a gel. Biodegradable plastic is what pectin is classified as Codex Alimentarius, a non-profit organization that sets global food safety standards, has determined that pectin is safe for human consumption. The Codex General Standard for Food Additives covers pectin, but there is no recommended maximum daily intake (IPPA, 2008). Because of its natural abundance, low cost, and renewable nature, pectin has been selected as one of the most essential raw materials for manufacturing soluble films. It was a huge success innovation to replace the synthetic film that is not environmentally friendly due to its biodegradable nature. When used as a film, pectin is an excellent barrier to oxygen, aroma preservation, and oil barriers, but it must be limited. Because pectin is a hydrophilic polymer soluble in water, it has a water limiting property Tulimandi et al. (2016) created a biodegradable and edible packaging film using papaya puree, gelatin, and defatted soy protein.

Table 2.1: List of reference on current study of pectin films

No.	Author /Year	Title Article	Details
1.	Liu et al., (2020)	Heat sealable soluble soybean polysaccharide/gelatin blend edible films for food packaging applications	The study's objective was to improve heat sealable soybean polysaccharide (SSPS)/gelatin blend films for use as edible food packaging. SSPS/gelatin blends were used to make the films, which were then plasticized with glycerol using a solution casting method. The results showed that combining SSPS and gelatin significantly improved the films' heat sealability, stretchability, and fracture resistance. Gelatin improved the thermal stability of SSPS films while lowering their water solubility, rigidity, and water vapour permeability. ATR-FTIR spectra and DSC results revealed strong interactions between SSPS and gelatin. Optical characteristics, SEM, AFM, and XRD tests demonstrated that SSPS and gelatin in this blend system were to some extent compatible. Packaging testing demonstrated that the blend films might be used as an edible material in food packaging.

2.	Tulamandi et. al., 2016	A biodegradable and edible packaging film based on papaya puree, gelatin, and defatted soy protein	Physical and mechanical properties of edible films made from different combinations of papaya puree (PP), gelatin (G), and defatted soy protein (DSP) were investigated. At room temperature, the films were cast using film forming solutions containing different amounts of papaya puree, gelatin, and defatted soy protein. The addition of gelatin to the papaya puree greatly improved the colour features, tensile strength, and seal strength of the films ($P<0.05$), according to the findings. The films' elongation, water permeability, and water contact angle all increased considerably ($P<0.05$) when defatted soy protein and gelatin were added to the papaya puree. DSC analysis also demonstrated that papaya puree blended films (PP/4DSP/3G) had a higher transition, melting temperature, and enthalpy than bleached papaya puree films (PP/4DSP/3G). Using Fourier transform infrared spectra analysis of blended papaya films, researchers identified the compatibility of papaya puree, gelatin, and defatted soy protein, as well as their influence on optical, mechanical, barrier, and structural properties. Papaya mixed films may soon be used as a crucial packaging material as a result of these results.

3.	Otoni et. al, 2017	Recent Advances on Edible Films Based on Fruits and Vegetables—A Review	Food packaging materials are usually assumed to preserve goods from degradation agents while also containing them. In addition to fruits and vegetables, polysaccharides and polypeptides provide edible packaging with acceptable physical-mechanical capabilities as well as different sensory and nutritional characteristics. This article examines the evolution of films made from fruit and vegetable purees, pomaces, and extracts over time. Recent advancements are thoroughly examined, with a focus on the role of each film component in the final materials, whose manufacturing procedures are examined from a technical standpoint, and key attributes are compiled and presented. Finally, the benefits and drawbacks of edible films made from fruits and vegetables are discussed in this comprehensive analysis.

2.4 GLYCEROL

Only food-grade ingredients should be used in soluble films, including the film-forming matrix, solvent, plasticizers and other additives. Gelatin films, for example, offer excellent mechanical and gas barrier properties, but they are frequently too brittle to be used in practical applications, necessitating the use of plasticizers. External plasticizers, in particular, diminish polymer chain-to-chain contact by separating nearby chains by taking up space between polymer molecules. As a result of this action, materials have less brittleness and stiffness and more flexibility, stretchability, and toughness (Otoni et al., 2017). Glycerol is the most commonly used plasticizer for this purpose (or vegetable glycerin).

Polyols, mono-, di-, and oligosaccharides are the most commonly used plasticizers. Polyols include glycerol, which has a high hygroscopic molecule and is commonly used with film-forming solutions to prevent brittleness in films. Several studies have demonstrated that glycerol can be used to make polysaccharide-based films like pectin. By increasing the mobility of amylase and amylopectin chains, it provided effective plasticization and increased film flexibility. High glycerol concentrations, on the other hand, may reduce tensile strength while increasing solubility, moisture content, and pectin film elongation at break. Glycerol, a small polyol with three hydroxyl groups, has been discovered as the best plasticizer for water-soluble polymers.

2.5 PAPAYA PUREE

Fruit and vegetable post-harvest losses are extremely high in food chain systems. Overripening of fruits prior to consumption is a major cause of post-harvest losses. Among the fruits, papaya had the highest post-harvest losses, accounting for about 75% of total produce (Paull et al., 1997). Papaya has been identified as a plant with medicinal properties, and it is also known as a nutraceutical plant due to its multifaceted medicinal properties (Milind et. al., 2011). As a result, scientists looked into the prospect of making edible and biodegradable films out of papaya puree to improve food quality while reducing waste.

Referring from Altaf et al. (2018), the highest yield (16%) of pectin was obtained at pH 2.0, for extraction time 60 min and extraction temperature 80 °C, in case of HCl and for citric acid extracted pectin highest yield (9.9%) was obtained at pH 2.0, for 60 min time of extraction and extraction temperature 80 °C. Pectin is found in papaya puree and can be used to generate biodegradable, environmentally friendly edible films (Tharanathan, 2003). Sugars, pectin, and a few protein molecules make up the papaya control films. The most important component in papaya is pectin, a complex heteropolysaccharide composed primarily of shifting galacturonic acid and rhamnose residues, as well as some arabinan and potentially galactan side chains. Their primary building blocks, on the other hand, are a-D-galactopyranosyl acid units and their methyl ester derivatives (Sila et al., 2009).

Functional chemicals in fruit and vegetable-based soluble films may improve the sensory, nutritional, and/or microbiological aspects of the packaged product or the packaging material itself, interacting with foods and playing an active role (Otoni et al., 2017). Papaya fruit was used in this study because it is high in provitamin A, ascorbic acid, B complex vitamins, and phytochemicals with antioxidant properties (see Table 2.1). According to Jabatan Pertanian Pulau Pinang (2020), in Malaysia there are three papaya varieties which are Exotica, Exotica 11, and Sekaki/Panjang. When papaya puree was added to pectin films, the tensile strength and elastic modulus were reduced, according to Otoni et.al. (2014). The elongation at break and WVP, on the other hand, were both increased, suggesting that the puree had a plasticizing effect.

Table 2.2: Nutritional value of papaya fruit (Source: Milind et. al., 2011)

Elements	**Amount (per 100 g)**
Energy	163kJ(39kCal)
Carbohydrates	9.81 g
Sugar	5.90 g
Dietary fibre	1.8 g
Fat	0.14 g
Protein	0.61 g
Vitamin A	55 µg (6%)
Beta-carotene	276 µg (3%)
Thiamine (Vit. B_1)	0.04 mg (3%)
Riboflavin (Vit. B_2)	0.05 mg (3%)
Niacin (Vit. B_3)	0.338 mg (2%)
Vitamin B_6	0.1 mg (8%)
Vitamin C	61.8 mg (103%)
Calcium	24 mg (2%)
Iron	0.10 mg (1%)
Magnesium	10 mg (3%)
Phosphorus	5 mg (1%)
Potassium	257 mg (5%)
Sodium	3 mg (0%)

Percentages are relative to US recommendations for adults

2.6 WATER SOLUBILITY, WATER VAPOUR PERMEABILITY (WVP), MOISTURE CONTENT

Water solubility is one of the most essential features of soluble films. Solubility refers to the ability of soluble films to dissolves in water for proper digestion when consumed, or it can naturally decompose when discharged into the environment. The film produced during this study is expected to achieve high solubility. It is intended to pack agar-agar powder which will be soluble when immersed in water to reduce packaging waste.

A portion of soybean polysaccharide (SSPS) combined with gelatin was immersed in 50 mL of distilled water in a study published in the journal by Liu et al., (2020). A higher percentage of SSPS on a gelatin-based matrix was found to result in greater film solubility. Blended films, on the other hand, lost water solubility as the weight fraction of gelatin increased. This is because gelatin aids in the formation of more compact polymeric matrices, allowing for a reduction in blend film-free volume with a higher gelatin content. Because both gelatin and pectin are thickening agents, pectin-based film incorporating papaya puree is expected to have higher water solubility than pectin film.

Furthermore, soluble film performance in food packaging applications is influenced by water vapour permeability (WVP) and water vapour transmission rate (WVTR). The film's WVP increases as the WVTR rises. The WVP value of the papaya puree pectin-based film is expected to be lower than that of the pectin film. Incorporating fruit macromolecules into the polymer matrix, according to Alves et al. (2011), can improve the vapour barrier properties. The dry cup method is the most often used method for determining a film's WVP since it is inexpensive and straightforward. Liu et al. (2020) also used a similar approach

The moisture content of the soil refers to the amount of water present in the soil (sample). The percentage ratio is calculated by dividing the quantity of water in a sample by the mass of solids in the sample. A moisture analyzer can be used to determine the moisture content of the powder because it is quick and affordable. The moisture content is strongly influenced by the WVP and WVTR of the type of film used for food packing. As a result, pectin-based film incorporating papaya puree is expected to have lower moisture content in agar-agar powder than pectin film, which has a higher WVP. According to Alves et al. (2011) and Liu et al. (2020), incorporating fruit and vegetable macromolecules would improve the stability of moisture barrier ability.

2.7 TENSILE PROPERTIES AND HEAT SEAL STRENGTH

The tensile properties of films are important for maintaining the mechanical integrity of food packages. Tensile strength (TS), elongation at break (EAB), and Young's modulus (YM) measurements are required to compare the strength of both types of films to develop a sustainable soluble and soluble film. A texture analyzer based on the standard D882-02 is one of the most common methods for determining tensile strength (ASTM, 2002). It is a texture measurement system that compresses or stretches a sample in either an up or down direction, making it ideal for determining the TS and EAB of film. Several studies on measuring the TS and EAB of biopolymer films, such as Liu et al., (2020), have used a similar method.

To prevent product leaking inside the package during storage, processing, or handling, the packaging material must have appropriate seal strength. According to Liu et al. (2020), a major disadvantage of soluble films is that they have poor heat seal properties. According to preliminary research, melting and fusion of pectin-based and papaya puree pectin based films could be achieved by using heat seal level 8 of the impulse heat-sealer Model SP-300H and heat sealing the film strip three times (Nur et al., 2020). According to Liu et al. (2020), a major disadvantage of soluble films is that they have poor heat seal properties.

2.8 ATOMIC FORCE MICROSCOPY OF BIO-BASED FILM

Atomic force microscopy (AFM) is a popular technique for producing topographical pictures of a sample surface by scanning the cantilever across a region of interest. The deflection of the cantilever is influenced by the peak and trough characteristics on the sample surface, which is monitored by the position-sensitive light diode (PSPD). Thus, AFM enables researchers to study the surface roughness of the samples, which is suitable for analyzing the surface roughness of a film. Liu et al. (2020) also performed the same method for analyzing the surface roughness.

According to the AFM results, the addition of gelatin, which caused tiny clusters to develop on the film's surface when the weight excess of soybean polysaccharide (SSPS) was too considerable, influenced the shape of the resulting film, according to Liu et al. (2020). Because both gelatin and pectin are thickening agents, pectin-based film incorporating papaya puree is expected to have a rougher film surface than pectin film. Moreover, the addition of papaya puree in pectin film will thicken the film, thus expected to increase the surface roughness.

2.9 OPTICAL PROPERTIES

In practical food packaging applications, colourless and transparent film packaging is preferred. The most appropriate and widely used method for analyzing optical properties is colorimeter. Colorimetry can generate L*, a*, and b* values, which are used to determine the colour pigment of a sample and the total colour difference. Liu et al. (2020) used a similar method to investigate the optical properties of biopolymer-based films. The film's transmittance, which will be used to determine transparency and opacity, is frequently measured with a spectrophotometer.

Transparency and opacity values are nearly identical. In terms of meaning, however, they are diametrically opposed. The lower the transparency value, the less transparent the film is, indicating it is opaquer. Total colour difference, as defined by Pathare et al. (2013), is the magnitude of the colour difference between stored and control samples (standard plate). Analytically, it can be divided into three groups: very distinct ($\Delta E>3$), distinct ($1.5<\Delta E<3$), and small difference ($1.5<\Delta E$).

2.10 FILM APPLICATION

The pectin film and papaya puree pectin-based film developed for this study were designed to pack 3 g of agar-agar powder in a small batch. This packaging pouches were made in accordance with (Liu et al., 2020). One of the most notable advantages of edible packaging systems over synthetic packaging systems is that they are an intrinsic component of the food product; they may be consumed without unpacking and discarding the package. The ability of edible films to dissolve in water so that they can be properly digested when consumed or decompose naturally if released into the environment is referred to as solubility. Soluble-film packaging is ideal for use in ready-to-eat food products since it melts in hot water or in the mouth of the consumer (Yai, 2008)).

Banana films produced by Sothornvit & Pitak, (2007) have good sealability, suggesting that they could be employed as dry food sachets or pouches. Thus, a pectin-based papaya puree film with good sealability can be used as a dry food sachet or pouch. Active substance carriers and controlled release are two of the functions and advantages of soluble film. Food components, pharmaceuticals, nutraceuticals, and agrochemicals can all benefit from soluble films and coatings in the form of capsules, microcapsules, soluble strips, flexible pouches, and coatings on hard particles (Suput et al., 2015).

2.11 AGAR-AGAR POWDER

The agar-agar powder used was for this study is Nona Serbuk Agar-agar. The ingredients composition in the agar-agar powder are 96% sugar, 3.25% agar-agar powder and 0.7% vanilla powder. The agar-agar powder needs to be packed with soluble pectin film for reducing the waste produce by using the synthetic polymer packaging. Also, this soluble packing packed with agar-agar powder can be dissolved in water and can be eaten without having to unpack and discard the package. Liu et al. (2020), published a paper in which they used soluble film to make heat sealable soluble soybean polysaccharide/gelatin blend edible films for food packaging applications.

2.12 TRUE DENSITY

The gas pycnometer is frequently used to determine the true density of powder (Pathare et al., 2013). This instrument uses gas displacement to determine the true density of a porous sample. The displacing medium is usually an inert gas like helium. After sealing the sample in a known-volume instrument chamber, helium gas is introduced to allow expansion into a second precision internal volume. The sample solid phase volume can be calculated using the pressures measured when the sample chamber is filled and then discharged into a second empty chamber. The true density of solid material is the density minus the volume of any open and closed pores. Yu et al. (1995) also used a similar method.

CHAPTER 3

METHODOLOGY

3.1 FLOWCHART FOR THE PROJECT EXPERIMENTS

Chapter 3 presents the raw materials and methods used in this project. The project experiments flow processes are shown in Figure 3.1. All preparation methods and tests procedures, test specimen, equipment and operating parameters used are explained in detail in Chapter 3. From the flowchart, the first objective of the study was accomplished by characterizing the soluble papaya puree pectin-based films and pectin-based film properties, including water solubility, tensile, seal strength, water vapour permeability, surface roughness and optical. Water solubility test, texture analyzer, water vapour permeation test, atomic force microscopy and colorimeter were performed for this purpose. The soluble papaya puree pectin-based films were made using a formulation ratio of 1:0.5:1 (pectin:glycerol:papaya puree). The formulation for soluble pectin film, (control) is 1:0.5 (pectin:glycerol). The second objective was accomplished by evaluating the quality changes on stored agar-agar powder. The agar-agar powders were stored for 39 days at ambient (28 °C) and chill (4 °C) temperatures. At selected storage time, moisture content, true density and colour of agar-agar powder were measured using moisture analyzer, gas pycnometer and colorimeter, respectively.

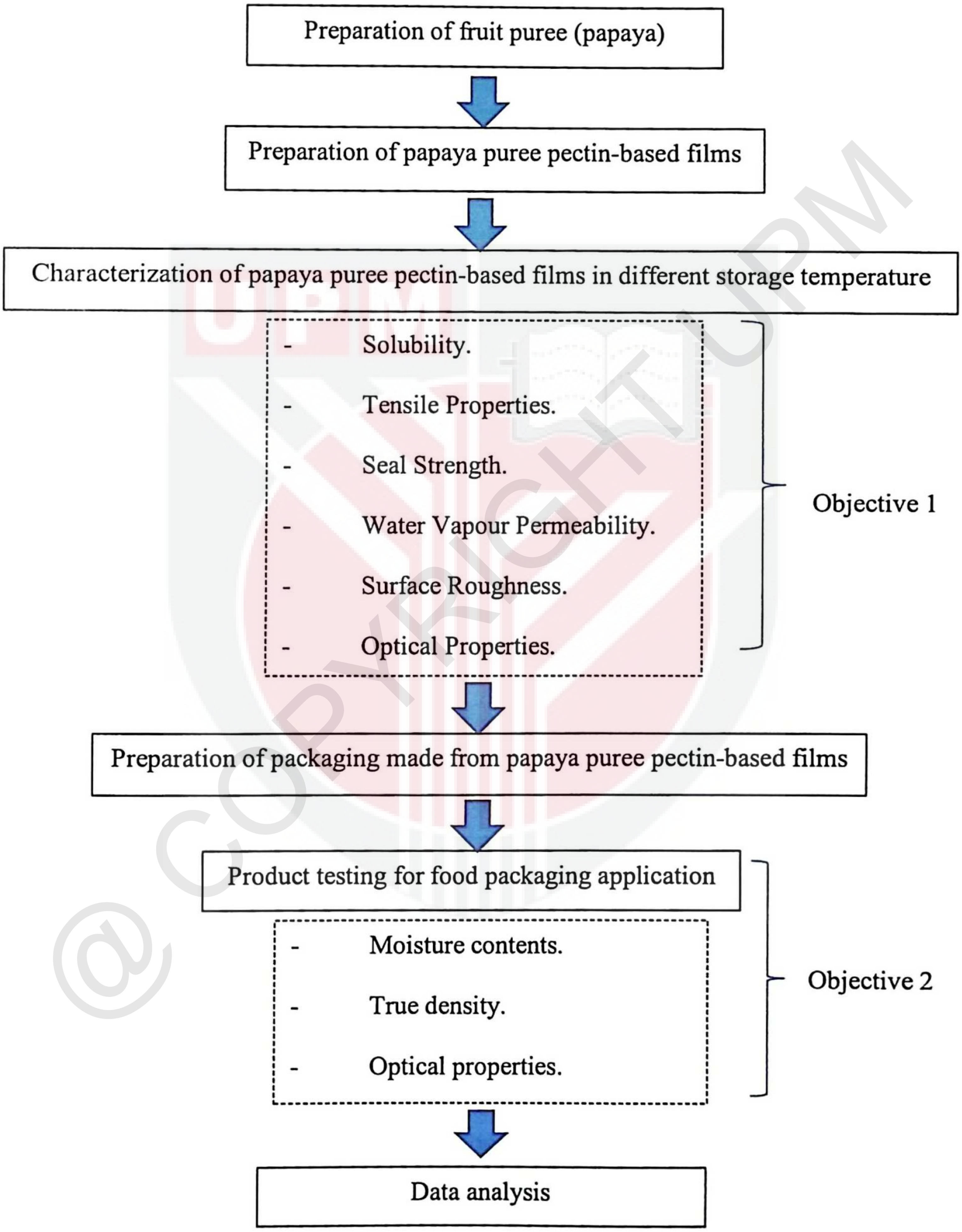


Figure 3.1: Flowchart for the project experiments

3.2 MATERIALS

Pectin (from R & M Chemicals, R & M Marketing, Essex, UK) and glycerol (from R & M Marketing, Essex, UK) were used to make the films (R & M Chemicals, R & M Marketing, Essex, UK). Papaya was bought from a wet market in MAEPS, Selangor. The papayas were picked at the peak of commercial ripeness and used as soon as possible after purchase.

3.3 PREPARATION OF PAPAYA PUREE

Before beginning the experiment, all of the apparatus were sanitized with a 70% alcohol solution. The fruits were thoroughly cleaned and washed before being air-dried. The fruits were peeled and then sliced into smaller pieces by hand. It was then blended using a food blender (MX-GM1011, Panasonic Co., Japan) until smooth. To obtain pure papaya puree, the blended mixture was filtered and rough particles were removed with a strainer. It was necessary to filter the papaya puree several times in order to obtain it. Fruit purees were then frozen and stored in a blue cap bottle with aluminium foil for later use.

3.4 PREPARATION OF SOLUBLE PECTIN-BASED FILMS

Two types of films were prepared: pectin film (control) and papaya puree pectin-based film. The formulation of the pectin film was according to (Maryam Adilah et al., 2018), the solvent casting technique prepared the films shown in Table 3.1.

Table 3.1: The formulation of films based on component ratio.

Films	Components	Ratio
Pectin film (control)	Pectin: Glycerol	1:0.5
Papaya Puree Pectin -based Film	Pectin: Glycerol: Papaya Puree	1:0.5:1

The film-forming solution was made by heating 100 mL of distilled water to 70°C and adding 4 g of pectin and 2 g of glycerol for 30 minutes. Using a magnetic stirring hotplate, the magnetic stir (04009951780017, DAIHAN Scientific Co, Korea) was kept at 100 rpm and 95 °C for complete cross-linking and gel formation pectin molecules. The papaya puree needs to be stirred using a smaller magnetic stirrer for 5 minutes to prevent a large clump of papaya puree for a uniform and smooth film. The solution was then cooled down to 50 °C before incorporating the papaya puree into the solution.

Then, for 5 minutes, the mixture of the film-forming solutions was stirred. The dissolved solution was then chilled for 1 hour at 4 °C in a chiller. Next, the mixture was sonicated for 10 minutes at 50 % amplitude using an ultrasonic processor for bubble removal. Finally, 20 mL of the solution was poured into glass Petri dishes and dried for 4 hours at 60 °C with no fan setting to ensure the films were dried completely. Before further analysis, the dried films were manually peeled from the Petri dish, cut to their proper shape, and stored in a drying cabinet at 30 °C with 50 % (RH) for 48 hours.

3.5 PREPARATION OF PACKAGE MADE FROM PECTIN-BASED FILM

A heat seal machine Model SP-300H. (Triumph Mercantile Corp, Taipei, Taiwan) was used to create a series of two-sided sealed semi-finished pouches (5 cm x 6 cm). Agar-agar powder (3 g) was manually transferred through the unsealed pouch's opening on one side. The pouches were then immediately heat sealed using the same seal condition. For the package test for food packaging application, sealed pouches containing agar-agar powders were stored for 39 days at ambient (28 °C) and chill temperatures (4 °C) for the study of the effect of the films on the quality changes in stored agar-agar powder. Figure 3.2 shows the sample of pouch filled with agar-agar powder used in the study.

Figure 3.2: Pouch prepared from pectin film and papaya puree pectin-based film

3.6 CHARACTERIZATION OF PAPAYA PUREE PECTIN-BASED FILM

3.6.1 WATER SOLUBILITY TEST

The percentage solubility of polymeric pectin films in water was determined using a water solubility test. With some modifications, the procedure analysis method was implemented using (Tulamandi et al., 2016) procedure. A razor blade was used to cut films into a square shape (4 cm x 4 cm) before testing the pectin. The films were then dried for 24 hours at 105 °C in a hot air oven until they reached a constant weight. The dried films were then placed in a desiccator containing silica gel at 0% relative humidity (RH) for 30 minutes. Following that, the films' initial dry weight was determined (W_i).

The dried films were then immersed in 50 mL of distilled water and left at room temperature for 24 hours in a Shaking Water Bath (Model BS-21, Jeio Tech Inc., Korea). After 24 hours of water immersion, the insolubilized films were filtered with filter paper (Whatman No. 541), dried, and weighed at 105 °C for 24 hours. Following filtration, the insoluble films were removed from the filter paper and dried in a hot air oven under the same conditions, yielding the final dry weight (W_r). Finally, the water solubility of each pectin film was tested in triplicate. The percentage of water solubility was calculated using the following equation:

$$\text{Water Solubility (\%)} = \frac{W_i - W_r}{W_i} \text{ X } 100$$

where:

W_i = initial dry weight of film (g),

W_r = final dry weight of pectin film (g)

3.6.2 TENSILE TEST

To determine the average thickness, the average value was taken from five random locations around each film. The thickness of the film was measured using a Digimatic micrometer (Mitutoyo, Model ID-C112PM, Serial No. 00320, Mitutoyo Corp., Kawasaki-shi, Japan). The tensile strength (TS) and elongation at break (EAB) of the films were measured using a texture analyzer (Stable Micro System, model TA. TXplus, Surrey, England) according to the standard method D882-02 (Liu et al., 2020). (ASTM, 2002). As shown in Figure 3.3, the films were cut into a long strip with dimensions of 15 mm x 100 mm (length x width). Before testing, they were conditioned for 48 hours in an environmental chamber (BH-TH_50, SH Guangpin (Bogong) Test Equipment Manufacturing Co.Ltd., Shanghai, China) at 25 ± 1 °C and 50 ± 5% RH.

A rectangular strip of each type of films was cut which was cut for the tensile test, clamped between two grips on fixtures before moving until the film failed. It enables the analyzer to produce a load extension graph, which can report overall strength, tensile strength, percentage elongation at break, and film toughness. Pneumatic grips with a rubber coating allow for precise control of gripping pressure. A 5 kN load cell with a 5 mm/sec crosshead speed was used for the test. The force (N) and elongation (mm) were measured at the point of failure. For the data analysis, three out of five results of films that broke in the middle of the strip were used to clarify the mechanical features of the film in food packaging, which are tensile strength (TS), elongation at break (EB), Young's modulus (YM) and toughness of the film.

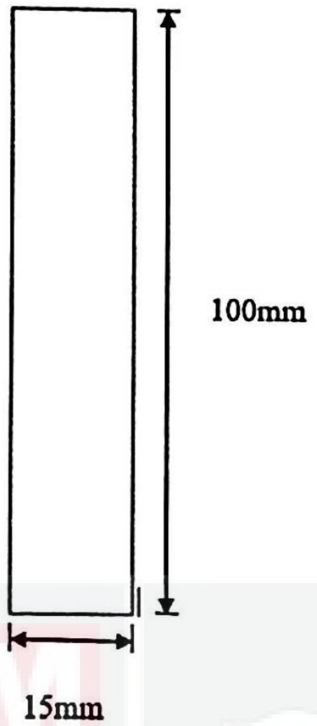


Figure 3.3: The example of cut film used for tensile test

The tensile strength of the films was obtained, where the tensile strength (TS) is the maximum strength of the sample under tensile loading. TS can be calculated before failure. Equation (3.2) expressing F_B is the force at breaking point.

$$\mathrm{TS} = \frac{F_B}{A_i} \tag{3.2}$$

Another test requires for mechanical strength is the elongation at break (EAB), which can be calculated as Equation (3.3), with L_B is the elongation at breaking point.

$$\%\mathrm{EAB} = \frac{L_B}{L_i} \times 100 \tag{3.3}$$

Young's modulus (YM), also known as modulus of elasticity, is the slope of the initial linear portion of the stress-strain curve, which can be calculated as follows:

$$\mathrm{YM} = \frac{\frac{\Delta F}{A_i}}{\frac{\Delta L}{L_i}} \tag{3.4}$$

where A_i is the film sample's first minimum cross-sectional area, L_i is the sample's primary length, Δ F and Δ L are the force and length changes.

3.6.3 SEAL STRENGTH TEST

Figure 3.4 shows how film samples were cut into 15 x 100 mm strips. Two strips were stacked on top of each other, and an impulse heat-sealer Model SP-300H was used to heat-seal a distance of 10 mm from the film's edge (Triumph Mercantile Corp, Taipei, Taiwan). The microprocessor kept track of the dwell time and temperature of each power level throughout the experiment. The film samples were then conditioned for 24 hours at 25 ±1 °C and 50% relative humidity (RH) before being used in additional tests to achieve chemical stabilization. As film strip samples, pectin film (control) and papaya puree pectin-based film were used.

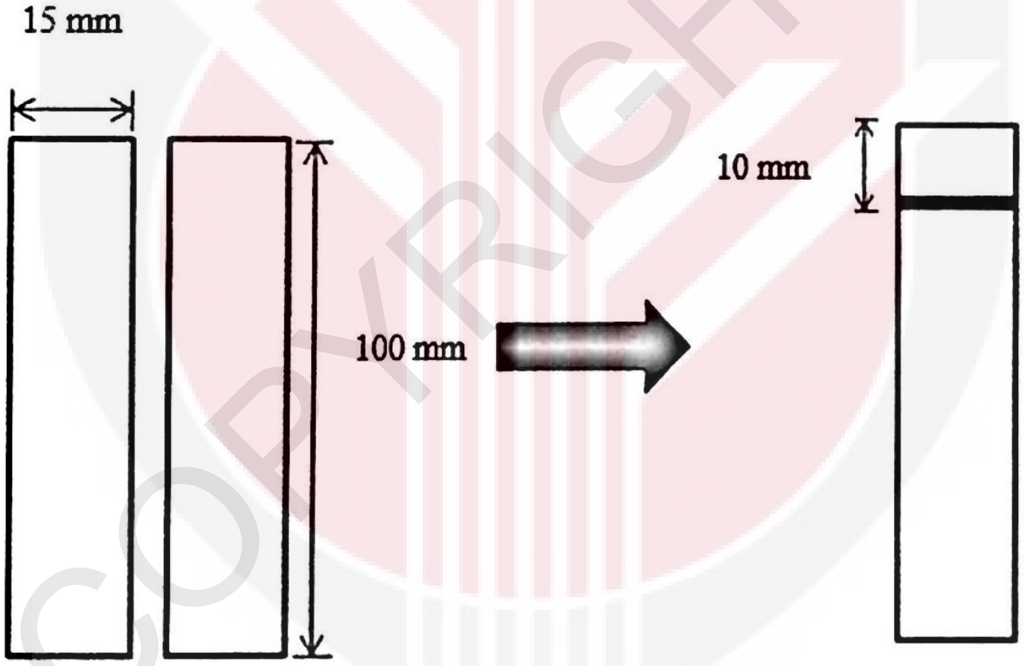


Figure 3.4: Fabrication of heat-sealed specimens and samples after cut and heat-sealed

A texture analyzer (Stable Micro Systems, model TA.TXplus, Surrey, England) and a T-peel test according to ASTM standard method D882-02 were used to determine the seal strength of the heat-sealed sample (ASTM, 2002). Each sealed film leg was clamped to the texture analyzer, and the sealed film's end was kept perpendicular to the pull direction, as shown in Figure 3.5. Then, at a constant rate of 200 mm/min, set the load. Using an unsupported head film technique consistently throughout the test helps to avoid inconsistent results.

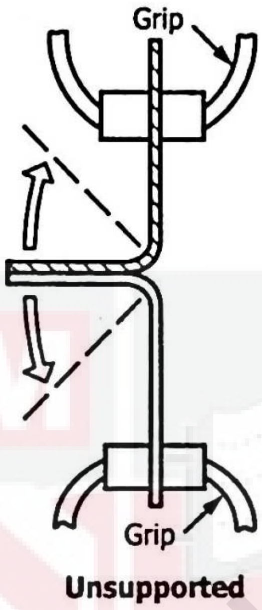


Figure 3.5: Overview of T-peel test of heat-sealed samples

3.6.4 WATER VAPOR PERMEABILITY (WVP) TEST

The WVP of films was determined using a modified method of ASTM standard E96-05 (Liu et al., 2020). The films were cut into circular samples and sealed onto circular test cups (diameter 60 mm). In a small glass dish, calcium chloride ($CaCl_2$) was placed. The distance between the film and the surface of the $CaCl_2$ is set at about 1.5 cm. At ambient temperature (28 °C), the test cups were placed inside a desiccator over a Magnesium hydroxide ($Mg(OH)_2$) solution 50 % (RH). For a period of seven days, the weight of each cup was recorded at 24 hours intervals. Equation (3.5) was used to calculate the films' WVP value:

$$\mathrm{WVP} = \frac{WVTR \times L}{\Delta\, P \times A} \tag{3.5}$$

WVTR stands for water vapour transmission rate, which is calculated from the slope of a weight loss vs. time plot. P is the difference in water vapour pressure across the film (kPa), and A is the film's area (m^2). The replicate for this WVP test is two replicates for each film.

3.6.5 ATOMIC FORCE MICROSCOPY TEST

The 3D surface topography of representative films was recorded in tapping mode using a Keysight 5500 (N9410S, Keysight Technologies Canada Inc., Canada) atomic force microscope. All of the 256 x 256 pixel images were obtained by scanning square areas of size 5 m 5 m with a scan speed of 0.3 line/s (Liu et al., 2020). The AFM images, surface roughness (Ra), and root mean square roughness (Rq) of pectin and pectin papaya puree-based films were obtained using NanoScope analysis.

3.6.6 OPTICAL TEST

Transparency value

The colour of the film samples was measured using a HunterLab ColorFlex EZ45/0° colour spectro-photometer (HunterLab, Reston VA, USA), with D65 illuminant, 10° observer, in accordance with ASTM E308 and with reference to Siracusa et al (2018). The measurements were taken using the CIELab scale. Before the test, the instrument was calibrated with a black and white tile. Five random scenes from the films were chosen for analysis. The results are expressed using the L* (luminosity), a* (red/green), and b* (yellow/blue) parameters. Each sample's L*, a*, and b* values were averaged across ten readings, and the total colour difference (ΔE) was calculated using the equation below:

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \tag{3.6}$$

where ΔL^*, Δa^* *and* Δb^* are the differentials between a sample colour parameter and the colour parameter of a standard (L^*=100.15, a^*=-0.02, b^*=—0.07) used as the film background.

The following formula was used to determine the transparency value:

$$\text{Transparency value} = \log T_{600nm}/L \tag{3.7}$$

where T_{600nm} is the film specimens' transmittance at 600 nm, and L is the average thickness of the specimens (mm). The lower the transparency value, the higher the transparency of the films.

The opacity is the opposite of the transparency value. The opacity of the films was calculated by the following equation.

$$\text{Opacity} = A_{600nm}/\delta \tag{3.8}$$

where A_{600nm} is the value of absorbance at 600 nm and δ is the film thickness in mm. For calculating the absorbance at 600 nm, it can be calculated from percent transmittance (%T) using this formula:

$$\text{Absorbance} = 2 - \log(\%T) \tag{3.9}$$

Transmittance is the percentage of incident light that is transmitted (T). Simply put, the amount of light that "successfully" passes through and emerges from the other side of a substance. The inverse of transmittance, absorbance (A), indicates how much light was absorbed by the sample. As a result, the higher the transparency value, the greater the opacity of the films.

3.6.7 STATISTICAL METHOD

All of the tests were carried out in triplicate. Data obtained were analysed using Minitab Statistical Software Version 16 (Minitab for Windows and Mac, Minitab inc., State College, Pennsylvania, USA) with a significance level of $p < 0.05$ used for mean comparisons. In addition, different treatments were statistically analysed for group differentiation using Tukey's analysis.

3.7 QUALITY CHANGES OF AGAR-AGAR POWDER PACKED WITH PECTIN-BASED FILM.

3.7.1 MOISTURE CONTENT TEST

Moisture content measurement was done using a moisture analyzer (Model MS-70, A&D Instrument LTD, Japan). The program used was set to 4 and the sample weight is 2 g. The analyzer first was equilibrated to make sure the reading obtained is correct. Then the agar-agar powder obtained from the pouch made from pectin film and papaya puree pectin-based film was weighed to 2 g on the metal petri dish, and its moisture was analyzed at 105 °C. The equipment will signal when the process is done and the result of moisture content will be obtained in percentage (%). The step was then repeated for each pouch containing agar-agar powder at ambient temperature (28 °C) and chill temperature (4 °C). Thus, each sample from each storage temperature has a duplicate, and readings were taken three times each.

3.7.2 TRUE DENSITY TEST

Micromeritics AccuPyc II 1340 Gas Pycnometer (Series #676, One Micromeritics Dr., Norcross, Ga 30093-1877 U.S.A.) was used for the measurement. True density refers to the mass of a unit volume of a solid in the air, minus any pore volume, permeable or impermeable. This value must be measured by passing a helium medium through all pores that does not interact with the substance. The analysis involves a helium gas used to displace the amount of the powder particle in terms of volume.

In this method, the volume and true density of the powder was obtained. The agar-agar powder from the pouch made from pectin film and pectin papaya puree-based film was filled ¾ into the 3.5 cm^3 cylinder for density measurement. The weight of the empty cylinder was weight, then filled in the agar-agar powder into the cylinder to obtain the sample weight. It was then placed inside the gas pycnometer, and the sample data were input into the software. The step was then repeated for each pouch containing product at ambient temperature (28 °C) and chill temperature (4 °C).

The data analysis was performed when 10 readings of the volume and the true density were obtained at the end of the analysis. The average of the readings was also given. Thus, the sample's true density can be calculated by dividing the average of the three volumes measurements into the sample weight in grams:

$$D = G/x \tag{3.10}$$

where D = sample density, G = sample weight (g), and x = average sample volume in cm^3.

3.7.3 OPTICAL TEST

Colour measurement of the agar-agar powder inside the pouch made from either pectin film and papaya puree pectin-based film was determined using a Precise Color Reader (Model WR-18, Shenzhen Wave Optoelectronics Technology Co., Ltd., China). The CIELab colour parameters were used for determine the accurate measurement and comparison of all perceivable colors using three color values. To execute the test, the device was first calibrated using standard white tiles, and the reading was taken at least three times.

Then, after constant reading of the white tiles was done, about 2 g of the agar-agar powder was placed on the white tiles for the colour measurement. The measurement was also done in triplicate. L^*, a^* and b^* values were averaged from three readings across for each sample and total colour difference (ΔE) was calculated according to Equation (3.6). ΔL^*, Δa^* and Δb^* are the differentials between a sample colour parameter and the colour parameter of a standard (L^*=88.35, a^*=0.44, b^*=1.51) used as the standard.

CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 CHARACTERIZATION OF PAPAYA PUREE PECTIN -BASED FILM

4.1.1 WATER SOLUBILITY

The water solubility of papaya puree pectin-based film is relatively low (9.2%) from the pectin film, as shown in Figure 4.1. Puree reduces the solubility of the pectin film because it is present. The extracts' phenolic chemicals, anthocyanins, vitamin C, and carotenoids (hydrophobic substances) disperse in the pectin films, lowering the polymeric structure's free volume (Eça et al., 2015). Table 4.1 shows the water solubility test results obtained for both pectin film and papaya puree pectin-based film.

Table 4.1: Solubility value for pectin film and papaya puree pectin-based film.

Type of Films	Solubility (%)
Pectin Film	93.7±5.8^{a}
Papaya Puree Pectin-based Film	84.5 ± 5.2^{a}

For each film, the data are reported as mean SD (n=2). Two columns with different letters (a, b) have significantly different mean values (p <0.05).

In addition, water vapour permeability (WVP) decreases in blend films as water solubility decreases. This could be explained by the fact that papaya puree aids in the production of more compact polymeric matrices, allowing the free volume in the blend film to decrease, resulting in lower WVP values (Liu et al., 2020). According to the results, papaya puree pectin-based film achieved similar water solubility values as mention in Tulamandi et al., (2016), where the water solubility of control papaya films is 77.54% ±1.04. The water-soluble pectin compound found in papaya films contributes significantly to the WS. The different in WS obtained for papaya puree pectin-based film could be due to the different composition of the film. According to Tulamandi et al. (2016), the papaya films were particularly hygroscopic in nature due to the pectin concentration in the papaya puree, which was shown to be readily wettable in water. In comparison to the pectin films, papaya puree pectin-based film had shown not significantly different (p > 0.05) water solubility (WS).

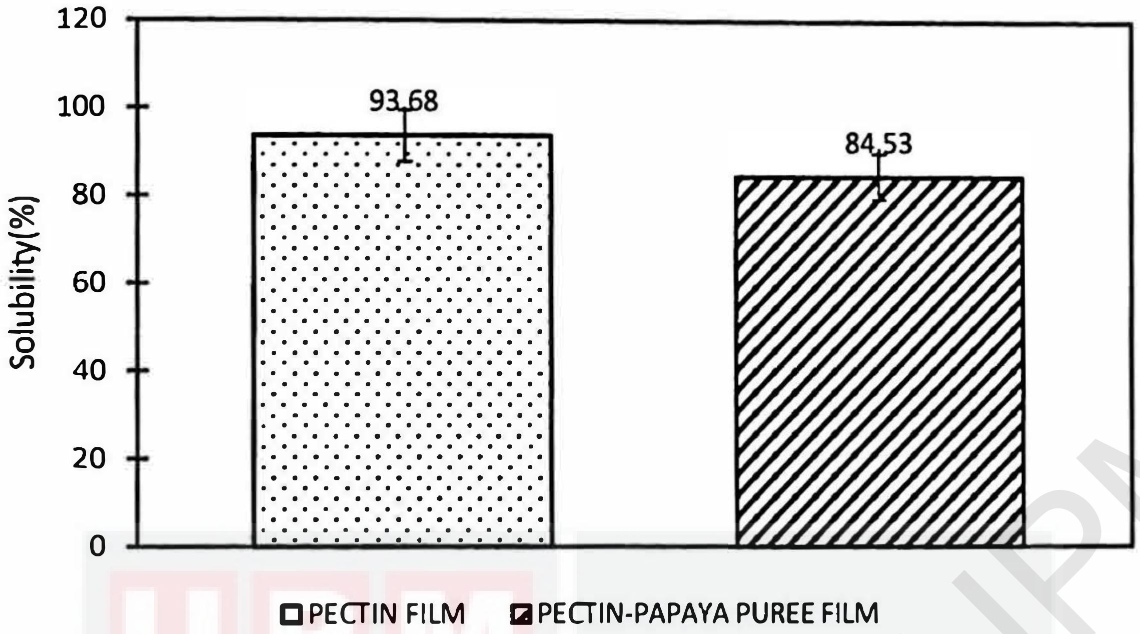


Figure 4.1: Water solubility (%) of Pectin film and Papaya Puree Pectin -based Film

4.1.2 TENSILE PROPERTIES

Films must have mechanical strength and flexibility in order to maintain the integrity of food packages. Table 4.2 shows the tensile strength (TS), elongation at break (EAB), Young's modulus (E), and toughness of pectin and papaya puree pectin-based films. The maximum stress that a material can withstand is measured in tensile strength. Elongation is determined by applying tensile force to the material or stretching it in the same way as previously described, then measuring the difference in length from the original. Young's modulus (E) measures a material's elasticity, which is defined as the relationship between a material's deformation and the force required to deform it.

The thickness of film used in the study is ranging from 0.1 until 0.08 mm. The thickness for pectin film is 0.085 mm while for the papaya puree pectin-based film is 1.0568 mm. The value of average thickness obtained from five reading.

Table 4.2: Tensile Strength (TS), Elongation at Break (EAB), Young's Modulus (E) and Toughness of Pectin Film and Papaya Puree Pectin-based Film

Type of Films	Tensile Strength (TS) (MPa)	Elongation at break (EAB) (%)	Young Modulus (E) (MPa)	Toughness (MJ/m^3)
Pectin Film	4.3 ± 0.1^a	42.1 ± 1.2^a	0.19 ± 0.01^a	1.07 ± 0.0^a
Papaya Puree Pectin-based Film	4.8 ± 0.4^a	54.9 ± 3.7^a	0.17 ± 0.04^a	1.77 ± 0.2^a

For each film, the data are reported as mean SD (n=2). Two columns with different letters (a, b) have significantly different mean values ($p < 0.05$).

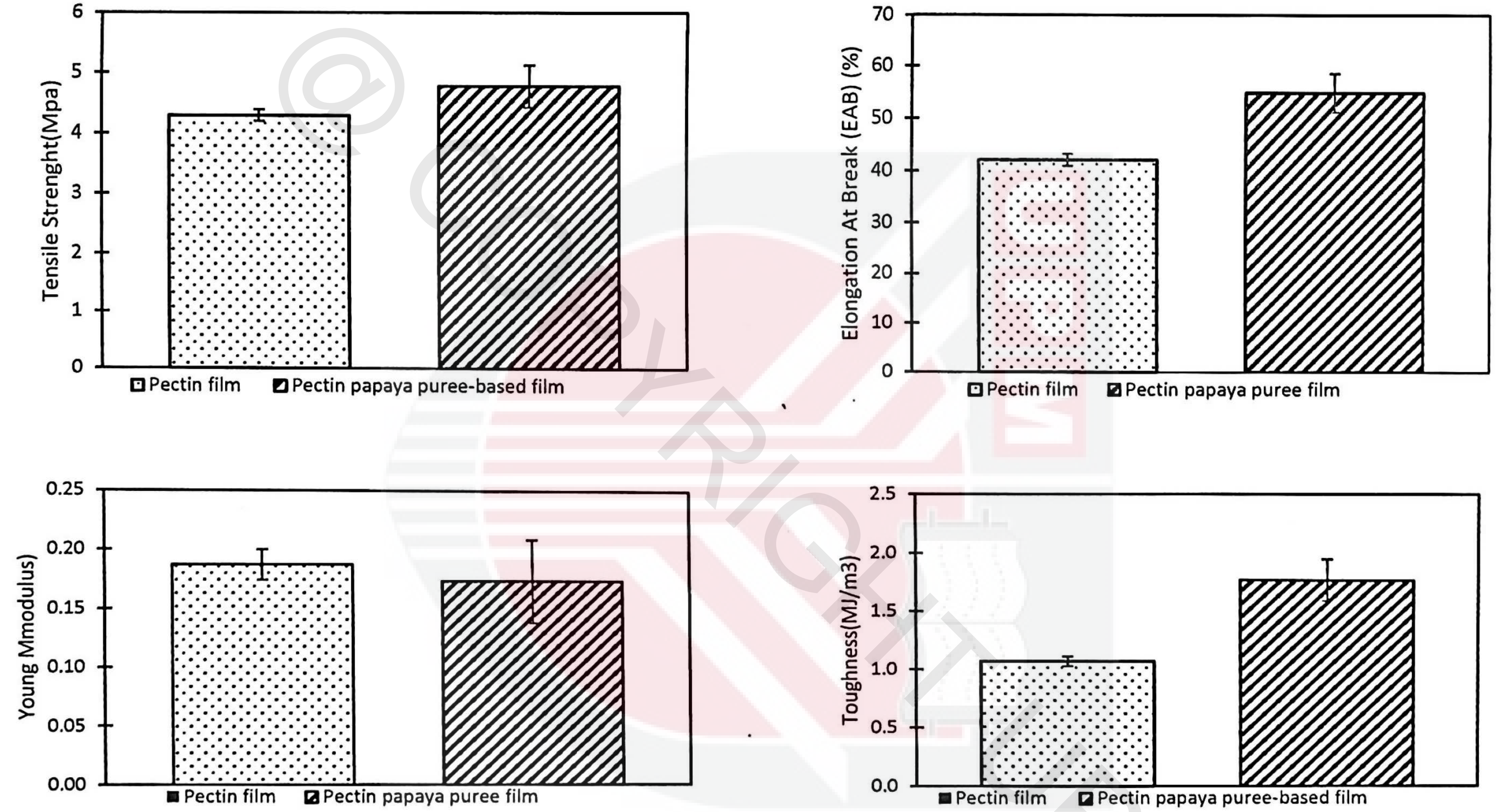


Figure 4.2:(a) Tensile strength (b) Elongation at Break (EAB) (c) Young's modulus (d) Toughness, of Pectin film and Papaya puree pectin-based film

The tensile properties result of the pectin film and papaya purees pectin-based film are shown in Table 4.2. Pectin film (control) shows a lower TS value with only 4.3 MPa ± 0.1[a], as shown in Figure 4.2 (a). However, when papaya purees are added into the film-forming solution, the TS value shows slightly higher than pectin film with 4.8 MPa ± 0.4[a]. The intense contact between the polymer and the fruit purees caused a cross-linking effect, which boosted tensile strength (Tulamandi et al., 2016). The findings were compared with several previous studies that noticed an increase in film strength after incorporating fruit purees. Edible films made from fruits and vegetables have tensile strengths ranging from 0.03 to 30 MPa, with papaya fruit with pectin binding agent having a tensile strength of 4.98-8.36 MPa at 33% (RH) (Otoni et al., 2017). Thus, the papaya puree pectin-based film results for tensile strength are in the range as mention.

Shown in Figure 4.2 (b), pectin film incorporated with papaya puree shows higher elongation at break (EAB) value with 54.9 ± 3.7 while control film with 42.1 ± 1.2. The high value of EAB shows that papaya puree incorporated with pectin film implements a strong elasticity and flexibility of film due to the cohesiveness of polymer molecules. Moreover, the incorporation of fruit purees increased the film's elongation because of the rise in free volume inside the film structure. TS and EAB were typically linked with the film through the intermolecular interaction between them. Otoni et al. (2017) observed that the elongation at break and WVP was increased, indicating that the puree had a plasticizing effect. The elongation at break of edible films was reported to range from 1.8% to 217% based for fruit and vegetable, while for papaya fruit with pectin binding agent with 33% (RH) has a 13.9-24.6% of EAB value (Otoni et al., 2017). The results obtained for the study for EAB is higher compare to the result obtained for film made of papaya fruit with pectin as binding agent at 33% (RH). This is because, both films consist of different composition and stored at different (RH) The result obtained for this study is when the film in a drying cabinet at 30 °C and 50% relative humidity (RH). Because the RH gradients between the two sides of films are the driving force for water vapour permeation, valid comparisons are achievable when different samples are subjected to the same RH circumstances (Otoni et al., 2014).

The results show in Figure 4.2 (c) proposed that there is a possibility that pectin film became more flexible with the presence of papaya puree. The Young's modulus value decreased from 0.19 MPa ± 0.01 to 0.17 MPa ±0.04 with the addition of papaya puree in the pectin film. The resistance and rigidity of the films were reduced by using papaya puree (Otoni et al., 2014). Young's modulus was reduced due to the plasticizing effect of the papaya puree. The Young's modulus of a flexible material is low, and it can change shape dramatically. Plasticization and blending with other materials are required, according to Vieira et al. (2011). This is for producing biopolymers film with enhanced mechanical properties. Similarly, the Young's modulus of edible films made from fruits and vegetables varies between 0.003 MPa and 1.0 GPa, while 30 - 125.26 MPa are the elongation at break (EAB) value for papaya fruit with pectin binding agent in 33 % (RH) (Otoni et al., 2017). The results obtained for EAB is in the range of EAB for edible films made from fruits and vegetables.

Figure 4.2 (d) shows that pectin film became tougher with the presence of papaya puree. Toughness of film is important for the packaging application such as for heat seal process. Furthermore, materials with a high tensile strength and a high elongation at break have a higher toughness. In comparison to the pectin films, papaya puree pectin-based film had shown not significantly different (p>0.05) tensile test. Because of the plasticizing effects of short-chain carbohydrates, more fruit and vegetable content are expected to result in reduced mechanical strength and stiffness, as well as greater extensibility (Otoni and others 2014), although the reverse behaviour has been documented (Souza et al., 2012). The presence of starch, cellulose derivatives, pectin, and other fibers has been responsible for the latter outcome.

4.1.3 SEAL STRENGTH

The edible films' seal strength (SS) was tested to verify their sealability and appropriateness as a principal packaging material for holding the actual food. Papaya films have a similar SS to bilayered corn zein and soy protein isolate films, as well as whey protein isolate films (Cho et al., 2010).

Table 4.3 shows the seal strength for pectin film and papaya puree pectin-based film. Heat-seal level 8 of the impulse heat-sealer Model SP-300H, and heat seal the film strip three times provided sufficient melting and fusion of pectin-based and papaya puree pectin-based film (Nur et. al, 2020). During the heat-sealing process, the heat would melt the crystalline bio polymer on the surfaces of two films that were pressed together between the heating plates. When heat and pressure were applied, the interfacial interactions between the films formed a joint. Based on the surface chemistry of the sealing material(s), a hot junction was formed after cooling due to re-crystallization of the polymers (Tulamandi et al., 2016). A strong and reliable packing meets the criteria to withstand the storage condition over a long period.

Table 4.3: Seal strength for pectin film and papaya puree pectin-based film.

Type of Films	Seal strength (N/mm)
Pectin Film	0.4±0.0^a
Papaya Puree Pectin-based Film	0.5±0.0^a

The results are presented as means SD (n=2) for each film. The mean values of two columns with different letters (a, b) differ significantly ($p < 0.05$).

Pectin film infused with papaya purees has a higher seal strength value of 0.5 N/mm ±0.0 than 0.4 N/mm ±0.0 for pectin film alone, as shown in Table 4.3. This could be due to the heat-sealed samples' high accessibility to heat due to low fibrous conditions, allowing for high sealing conditions. As a result, pectin film containing papaya puree has higher seal strength than pectin film alone. Compared to the pectin films, papaya puree pectin-based film had shown no significantly different seal strength ($p > 0.05$).

Banana films with good sealability were developed by Sothornvit and Pitak (2007), who suggested that they could be used as dry food sachets or pouches. Thus, papaya puree pectin-based film with a good sealability can be used as dry food sachet or pouches. Tulamandi and colleagues (2016) developed papaya puree films with seal strengths of 127.3 to 726.0 N/m using starch, gelatin, or soy protein. As a result, the papaya puree pectin-based film's seal strength falls within the specified range with 0.5 N/mm or 500 N/m.

4.1.4 WATER VAPOUR PERMEABILITY

When food is exposed to a lot of moisture, it becomes more prone to spoilage. As a result, food packaging must have a good barrier against water vapour permeability (WVP). According to Alves et al. (2011), incorporating inert impermeable barriers into the polymer matrix can enhanced the vapour barrier properties. WVP is influenced by macromolecule mobility and free volume, as well as the films' integrity and hydrophilic–hydrophobic and crystalline–amorphous ratios.

Based from results obtained in Table 4.4, the water vapour transmission rate (WVTR) value of pectin papaya puree-based film was slightly but significantly lower than pectin films. This finding is consistent with the solubility results, which show that as the WVTR and WVP decrease, the solubility decreases. This could be explained by the fact that papaya puree aids in the formation of more compact polymeric matrix, allowing for a reduction in free volume in the mix film, resulting in lower water vapour permeability (WVP) values (Liu et al., 2020).

The decreased in WVP values upon adding more papaya puree in the pectin matrix from 115.5 x 10^{-8} g /m. day Pa ± 9.9^{a} to 116.3 x 10^{-8} g /m. day Pa ± 33.2^{a} would be advantageous for refining the stability of packed food products. This is crucial because the capacity to control moisture content inside food containers is dependent on good water vapour barriers (Alves et al., 2011) Due to the higher sugar content in the papaya puree, which acts as a plasticizer and lowers the film's capacity to inhibit moisture permeability, the papaya puree pectin film showed lower WVP values (Tulamandi et al., 2016).

Table 4.4: Water vapour transmission rate (WVTR) and water vapour permeability (WVP) test for pectin film and papaya puree pectin-based film

Type of Films	WVTR (g/day.m^2)	WVP (x 10^{-8}) (g./m^2.day Pa)
Pectin Film	297.5±25.5^a	115.5 ± 9.9^a
Pectin-Papaya Puree-based Film	238.2±68.0^a	116.3± 33.2^a

The results are presented as means SD (n=2) for each film. The mean values of two columns with different letters (a, b) differ significantly ($p < 0.05$).

Based on Figure 4.3, the weight changes for pectin film are higher than the value for papaya puree pectin-based film. This result obtained is significant with the result for the film's water vapor permeability (WVP). The pectin film's WVP value is higher than papaya puree pectin-based film; thus, pectin film is more permeable to water. Because the major role of soluble film or coatings is to inhibit moisture transfer between food and the surrounding atmosphere or between two components of heterogeneous food products, water vapour permeability should be as low as possible. This could affect the storage of the product in food packaging applications where its quality might deteriorate faster than those packed in papaya puree pectin-based film.

According to Otoni et al. (2017), a similar trend has been discovered, with the WVP of fruit and vegetable puree edible films having a substantially higher WVP value than the average WVP of synthetic polymer films. When different samples are subjected to the same RH conditions, accurate comparisons are possible since the RH gradients between the two sides of films constitute the driving force for water vapour permeation (Otoni et al., 2014). As a result, RH gradients should be taken into account while comparing WVP. The water vapour permeability of the papaya puree pectin-based film was not substantially different ($p > 0.05$) from the pectin films (WVP).

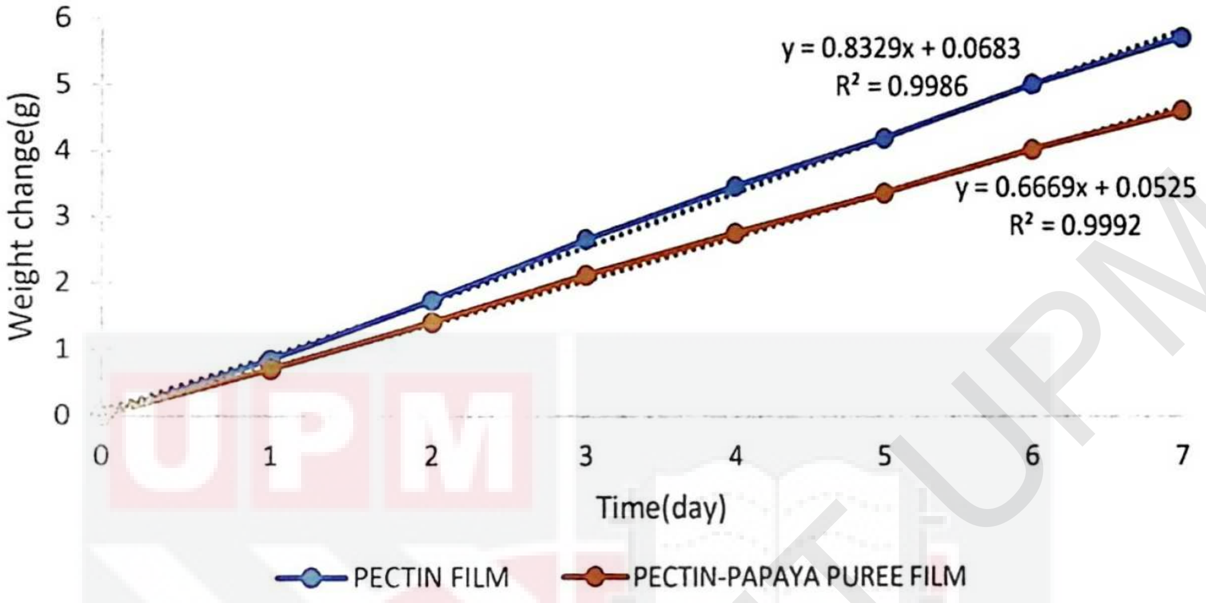


Figure 4.3: The weight changes of pectin film and papaya puree pectin-base film in 7 days

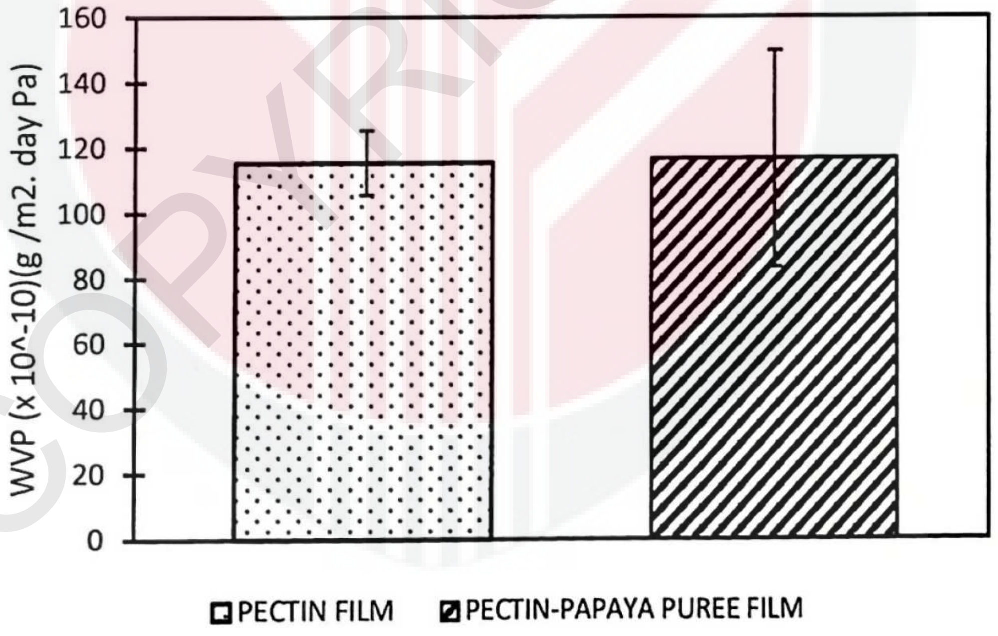


Figure 4.4: Water Vapour Permeability for pectin film and papaya puree pectin-based film

4.1.5 SURFACE ROUGHNESS

The results of atomic force microscopy (AFM) show the roughness of the film surface. Figures 4.1 and 4.2 show the AFM image of pectin and papaya puree pectin-based films' surfaces. The addition of papaya puree to the pectin film increased the roughness of the films significantly, as evidenced by the final values (after flattening) of average surface roughness (Ra) of pectin film and papaya puree pectin-based films of 5.2 nm and 6.9 nm, respectively. The root mean square roughness (Rq) also shows similar increasing trend with 7.1 nm for pectin film and 8.9 nm for papaya puree pectin-based film. Also, from Figures 4.5 and 4.6, the highest peak for papaya puree pectin-based film is higher with 32.3mm compared to pectin film with 30.3 mm, which also indicates a rougher surface of the film.

These results obtained supported by several journal that shows addition of fruit puree increase the surface roughness. According to (Go & Song, 2020.), insoluble particles and aggregates of rambutan peel extract (RPE) in the film matrix generate an uneven film surface. During drying, the insoluble RPE particles migrate to the film surface, resulting in a rough finish. The value of Ra and Rq for both film types revealed that the film incorporated with the papaya puree shows a rougher surface than the pectin film. In other words, the papaya puree diminishes the surface features of the film by increasing its roughness. Tulamandi et al. (2016) also discovered that adding gelatin to papaya films made the surface smoother and reduced the porosity nature of the films. The results obtained for papaya puree pectin-based film may not be a good quality for soluble packaging film as smoother surface are more desired.

Table 4.5: Value of Rq and Ra after flatten for pectin film and papaya puree pectin - based film

Sample	Image Rq (nm)	Image Ra (nm)
Pectin Film	7.1	5.2
Pectin-Papaya Puree -based Film	8.9	6.9

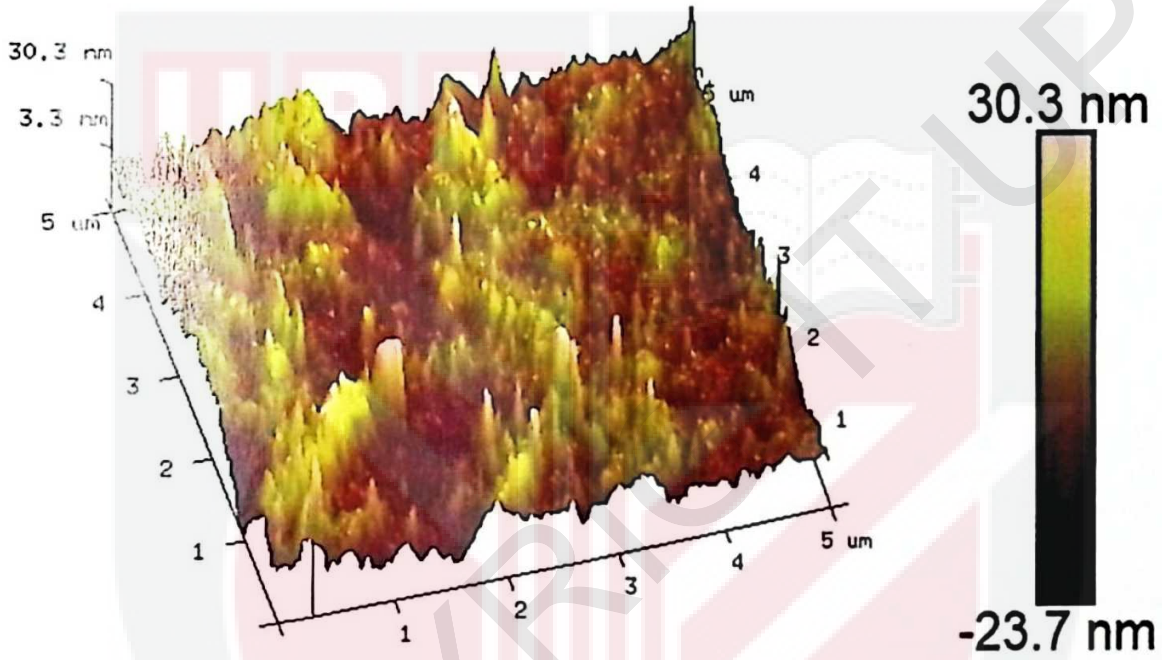


Figure 4.5: AFM image of pectin film

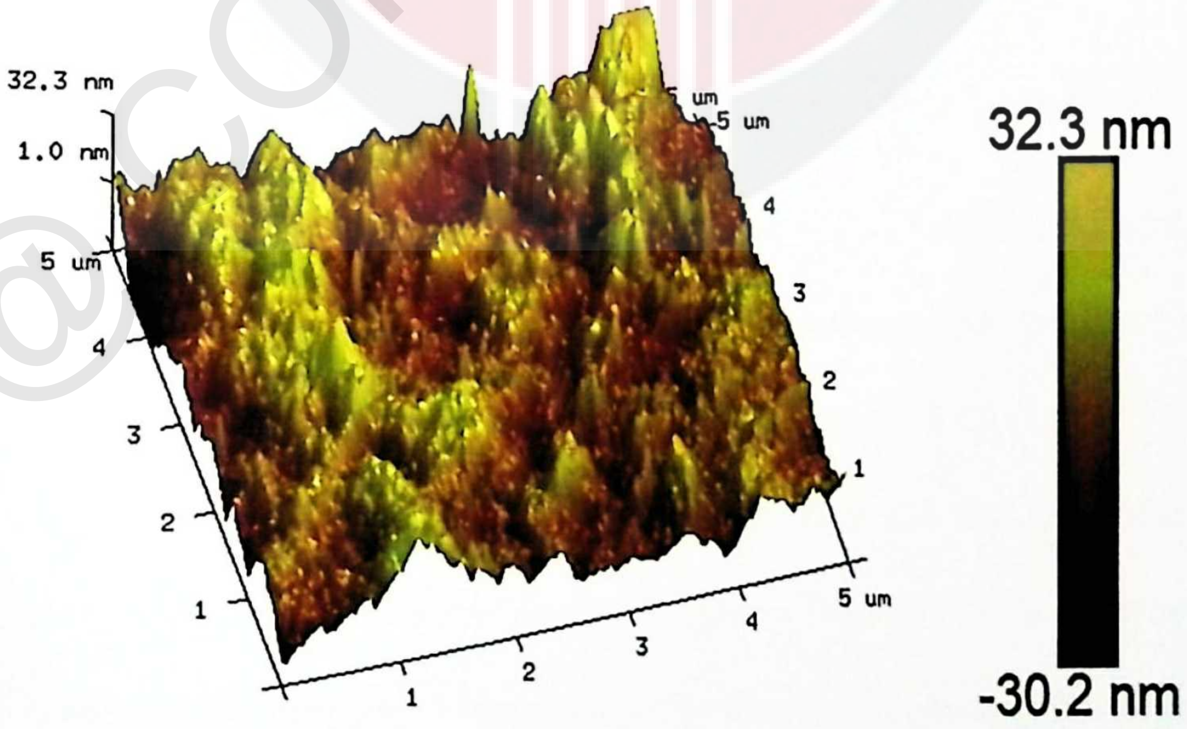


Figure 4.6: AFM image of papaya puree pectin-based film

4.1.6 OPTICAL PROPERTIES

Because it directly impacts on the appearance of the coated product, soluble film transparency is critical for consumer acceptance. The film should be colourless (or clear), translucent, and seem like polymeric packaging materials or the food colour to be coated. In film-forming emulsions, the oil volume fraction and droplet size distribution, as well as their rearrangement after drying, are studied. affect the internal structure of emulsion-based films. Thus, color parameters *L**, *a**, *b**, and film opacity were analyzed to understand better the optical properties of pectin films with papaya puree and the total colour difference (ΔE) of pectin-based film.

Table 4.6: Optical properties for pectin film and papaya puree pectin-based film

Film	L*	a*	b*	ΔE
Pectin Film	91.9 ± 0.3^a	-0.9±0.0^a	9.4 ± 0.3^a	13.0 ±0.3^a
Papaya Puree pectin-based Film	88.3 ± 0.3^b	2.2 ±0.2^b	19.4 ±0.5^b	22.8 ±0.0^b

The results are presented as means SD (n=2) for each film. The mean values of two columns with different letters (a, b) differ significantly ($p < 0.05$).

Table 4.6 shows the values of these optical parameters for all films studied. As can be seen, all of the films have high lightness (L*) values, which remained relatively constant after the addition of papaya puree, dropping from 91.9 ±0.3^a to 88.3 ± 0.3^b. L* stands for lightness, with 0 representing complete darkness and 0% reflectance or transmission. A higher L* value indicates lightness. Pectin film is more reflectance compared to papaya puree pectin-based film. These results are comparable to emulsified soluble films based on soy protein or gelatin (Nazmi et al., 2020).

The color's redness-greyness is represented by a*. a* values that are positive are red, while negative values are green. a* level of 0 is considered neutral. The a* value for pectin film, -0.9 ± 0.0, indicates a green tone, while 2.2 ± 0.2 for papaya puree pectin-based film indicates a red tone. The red tone indicates the presence of papaya puree in the pectin film b* indicates the color's yellow-blueness, with positive values indicating yellow and negative values indicating blue. Each film shows a positive b value which indicates the colour yellow. Thus, the colour of pectin film with the addition of papaya puree was less colourless with a slight yellowish red colour.

The total colour difference (ΔE) increased significantly from 13.0 to 22.8 when papaya puree was added. The results obtained throughout the storage period were compare to the colour parameter of agar-agar powder at day 0 ($L^*= 100.94$, $a^*=0.04$, $b^*=0.03$). The higher the value of ΔE of film, the greater the difference between the sample and the standard colours in that dimension. Therefore, the pectin film is closer to the standard compared to the papaya puree pectin-based film. In comparison to the pectin films, papaya puree pectin-based film showed a significantly different optical parameter ($p < 0.05$).

Table 4.7: Transparency and opacity value of Pectin film and papaya puree pectin-based film

Film	Transparency value	Opacity value
Pectin Film	0.92 ± 0.03^a	0.92 ± 0.03^a
Papaya Puree Pectin-based Film	0.85 ± 0.02^b	0.85 ± 0.02^b

The results are presented as means SD (n=2) for each film. The mean values of two columns with different letters (a, b) differ significantly ($p < 0.05$).

When papaya puree was added to pectin film, the transparency decreased from 0.92 to 0.85. The lower the transparency of the films, the higher the transparency value. Therefore, the pectin film is much clearer than papaya puree pectin-based film as the colour of papaya puree itself is bright orange colour with affect the transparency. The varying transparency levels should be linked to the variations in the internal structure development when many changes occur during drying (Galus & Kadziska, 2016). Anyway, the results of the transparency test reveal that both films are appropriate for food packaging. The papaya puree pectin-based films are more transparent than the commonly used commercial synthetic food packaging films such as low-density polyethylene (LDPE) (transparency value 4.26) and oriented polypropylene (transparency value 1.57) (Guerrero et al., 2011).

The opacity is the opposite of the transparency value. The presence of papaya puree in pectin film decreases the opaqueness from 0.92 to 0.85. This could be owing to the presence of a dispersed, non-miscible phase that promotes opacity due to refractive index discrepancies between the phases, as well as the dispersed phase's concentration and particle size (Galus & Kadziska, 2016). Therefore, the higher the opacity value, the opaquer the film is. When an element has a value of 0, it is completely transparent; when it has a value of 1, it is completely opaque (solid). Thus, the colour of pectin film with the addition of papaya puree was enhanced with the slight yellowish red colour but not as desired for the film to be colourless (or clear) and the film become less transparent and more opaque compare to pectin film.

4.2 QUALITY CHANGES OF AGAR-AGAR POWDER PACKED WITH PECTIN-BASED FILM.

4.2.1 MOISTURE CONTENT

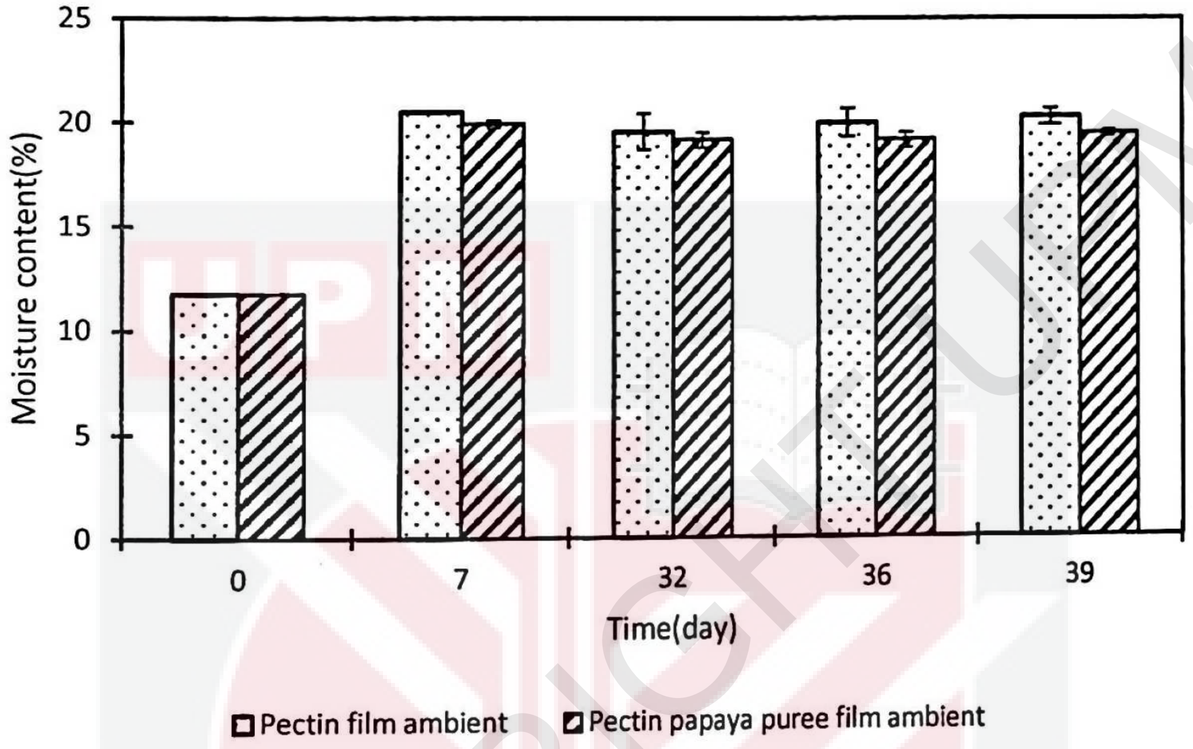


Figure 4.7: Moisture content of agar-agar powder in pectin film and papaya puree pectin -based film at ambient temperature storage

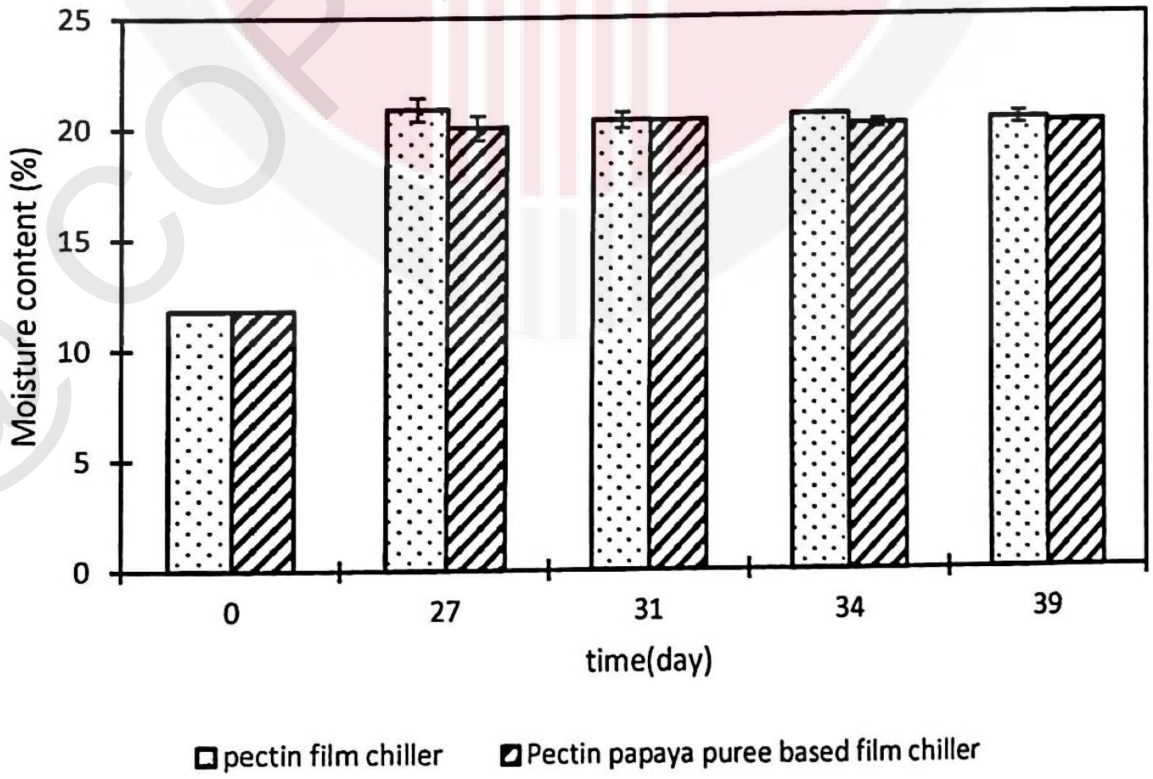


Figure 4.8: Moisture content of agar-agar powder in pectin film and papaya puree pectin -based film at chill temperature storage

Because efficient water vapour barrier ability is crucial for managing moisture content inside food packages, the drop in WVP values generated by the addition of papaya puree to pectin film would be useful for increasing the stability of packaged food products. Figures 4.7 and 4.8 show the moisture content of agar-agar powder ambient (28 °C, RH 50%) and chill (4 °C, RH 50%) respectively, from day 0 until day 39. The moisture content for the agar-agar powder is at 11.8% when measured from when it is store in the commercial synthetic food packaging. After 39 days in the pouch made from pectin film and papaya puree pectin-based film store in ambient temperature and chiller temperature, the moisture content for agar-agar powder in both films shows an increase in moisture content. These results indicate the agar-agar powder absorbed less moisture (water vapour) by using papaya puree pectin-based film pouches. This finding is supported by the value of WVP and WVTR of the papaya puree pectin-based film.

The result obtained related to the previous film analysis were the permeability of film with the addition of papaya puree increases film permeability (WVP of pectin film increase with addition of papaya puree from 115.5 x 10^{-8} g/m. day Pa ± 9.9 to 116.3 x 10^{-8} g/m. day Pa ± 33.2). The film barrier properties have improved due to matrix organization enhancement and obstructing water vapor passage through the film. Both papaya puree pectin-based film values were smaller than the pectin film, suggesting that papaya puree pectin-based film has better barrier properties. Thus, papaya puree pectin-based film is a better type of packaging for storing agar-agar powder in both ambient and chill temperature.

It can also be observed that the moisture content of agar-agar powder store at chill temperature has a higher increase in moisture content than the moisture content of product at the ambient temperature storage. This may be because the film absorbs the moisture present in the chiller more than in ambient temperature. The moisture content for the papaya puree pectin-based film in both ambient temperature and chiller temperatures is lower than the product in pectin film. Thus, it proves that the incorporation of pineapple puree in the pectin-based film makes the soluble film a better choice for food packaging application. According to Otoni et al. (2017), the fact that the puree is a hydrocolloid that acts as a binding agent with the film matrix improved the barrier qualities of the soluble film integrated with fruit puree.

4.3 TRUE DENSITY

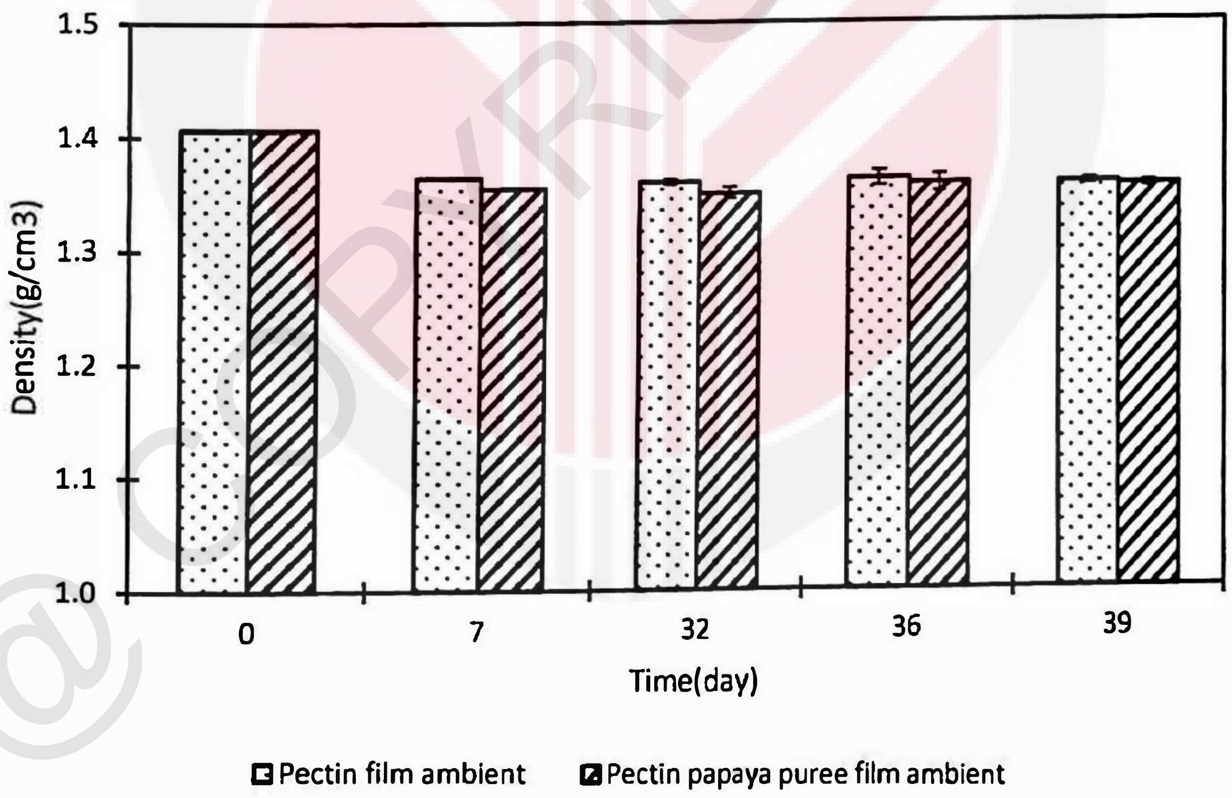


Figure 4.9: True density of agar-agar powder in pectin film and papaya puree pectin-based film in ambient temperature

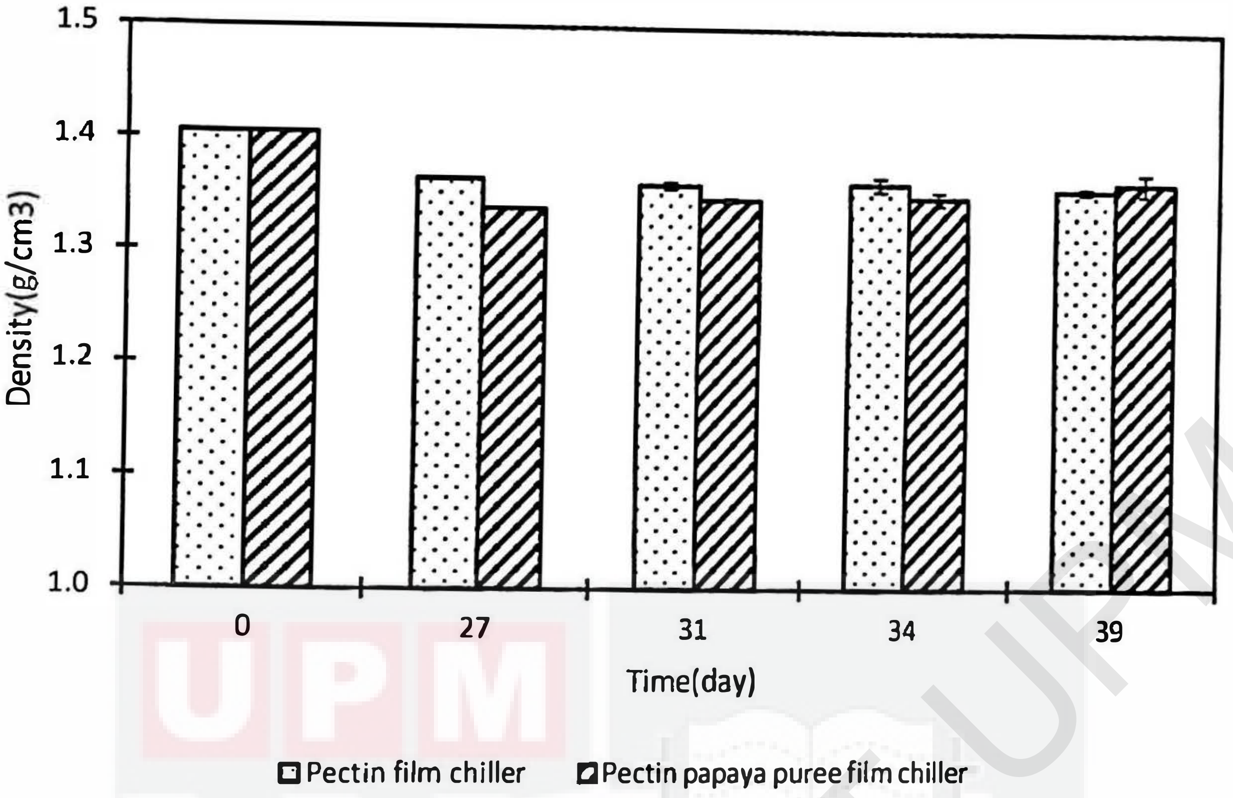


Figure 4.10: True density of agar-agar powder in pectin film and papaya puree pectin-based film in chill temperature

True density refers to the mass of a unit volume of a solid in the air, minus any pore volume, permeable or impermeable. This value is calculated by passing a medium through all pores that does not interact with the substance, commonly helium (Stanton, 1982). The true density measurement of agar-agar powder aids to determine the effect of the films on the quality changes in stored agar-agar powder, such as agglomeration of the powder. When water is added to agar-agar powder, agglomeration occurs, as the amount of agglomeration depends on how the powder is handled (Yu et al., 1995).

Figures 4.9 and 4.10 show the true density of agar-agar powder in ambient and chill temperature from day 0 until day 39. The true density of agar-agar powder at day 0 is 1.406 g/cm^3 when measured from when the agar-agar powder is store in the commercial synthetic food packaging. The true density for the agar-agar powder at day 39 is 1.3575 g/cm^3 for pectin film and 1.355 g/cm^3 for papaya puree pectin-based film for the ambient temperature. Both agar-agar powder in pectin film and pectin film with the addition of papaya puree shows a reduction in the density value than day 0.

However, the value of average density for pectin film at day 39 is higher compared to papaya puree pectin-based film as it related to the moisture intake more for the agar-agar powder in pectin film. The value of moisture content for agar-agar powder in pectin film is at an average of 18.6%, while for agar-agar powder in papaya puree pectin-based film is 18.2%. The agglomeration of powder particles, which increases as the moisture content rises, causes intraporosity. Because true density is defined as the density that excludes all open and closed pores, the agar-agar powder's true density decreased as the intraporosity caused by agglomeration of agar-agar powder particles increased. This theory is supported by the findings of a study on coal agglomeration due to moisture content (Yu. et al., 1995).

4.3.1 OPTICAL PROPERTIES

Tables 4.8 and 4.9 show parameters observed for pectin film and pectin papaya puree-based film in storage condition from day 0 until day 39 at ambient is 28 °C and RH 50%. Tables 4.10 and 4.11 on the other hand, show the results at a chill temperature from day 0 until day 39 at 4 °C and at RH 50 %. L* represents lightness. All of the films have high lightness (L*) values, which remained generally consistent until day 39, with a minor drop ranging from 94.3 ± 0.0 on day 0 to 86.7 ± 3.0 for the lowest value of L* for agar-agar powder in pectin film at ambient temperature. A higher L* value indicates lightness. According to Addala (2015), a longer period of storage and higher irradiation doses resulted in a significant ($p<0.01$) reduction in paprika powder carotenoids. Therefore, to have better understanding of the optical properties of agar-agar powder in pouches made from pectin films and papaya puree pectin-based film, colour parameters L*, a*, b*, and total colour difference (ΔE) were analyzed.

a* represents the redness-greyness of the color. Both agar-agar powder in pectin film and pectin papaya puree-based film has a positive value of a*, indicating a red tone. The range for a* value for both film types in ambient and chill temperatures is 2.6 ± 0.0 for the highest value and 1.3 ± 0.3 for the lowest value for agar-agar powder in pectin film at chill temperature. b* denotes the yellow-blueness of the color. Positive values of b* are yellow tone. Each film shows a positive b value which indicates the colour yellow. The range for b* value for both films in ambient and chill temperatures is 15.9 ± 0.4 for the highest value, and 12.1 ± 0.2 for the lowest value for agar-agar powder. The results obtained throughout the storage period do not shows an obvious change in the colour parameter than the colour parameter of agar-agar powder at day 0 (L*= 94.3, a*=1.4, b*=15.3). It is revealed that storage condition at chiller losses its colour intensity more than the one stored at ambient temperature for both type of films.

At ambient temperature, when agar-agar powder is in from pectin film, the total colour difference (ΔE) decreased significantly from 15.0 to 12.0. In contrast, agar-agar powder in a pouch made from papaya puree pectin-based film, the total colour difference (ΔE) decreased significantly from 15.0 to 10.8. On the other hand, at chill temperature, ΔE decreased significantly from 15.0 to 10.8 when agar-agar powder was in pectin film while the agar-agar powder in pouch made from pectin papaya puree-based film, ΔE decreased significantly from 15.0 to 11.9.

As mentioned above, the lower the value, the greater the difference in that dimension between the sample and the standard colour (white tile). The decrement of total colour difference (ΔE) on both storage temperatures and types of film indicates that the agar-agar powder loses its yellowish colour intensity during the 39 days storage time. Thus, agar-agar powder stored in pectin film shows less changes in colour (originally yellowish colour) compare to agar-agar powder stored in papaya puree pectin-based film. These findings contradict the results obtained in Liu et al., (2020), who found that due to the excellent transparency of the films in the visible range, all dry powders packaged in these pouches made from sealable soybean polysaccharide (SSPS)/gelatin retain their original colours.

Table 4.8: *Results obtained for the optical properties of agar-agar powder in pouch made from pectin film at ambient temperature.*

Day	L*	a*	b*	Total Colour Difference (ΔE)
0	94.3 ± 0.0	1.4 ± 0.0	15.3 ± 0.0	15.0 ± 0.0
32	88.5 ± 1.9	2.3 ± 0.7	15.9 ± 0.4	14.6 ± 0.4
36	86.7 ± 3.0	2.6 ± 0.0	14.0 ± 0.0	12.9 ± 0.3
39	90.1 ± 0.2	1.8± 0.0	13.3 ± 0.0	12.0 ± 0.0

Table 4.9: *Results obtained for the optical properties of agar-agar powder in pouch made from pectin papaya puree-based film at ambient temperature.*

Day	L*	a*	b*	Total Colour Difference (ΔE)
0	94.3 ± 0.0	1.4 ± 0.0	15.3 ± 0.0	15.0 ± 0.0
32	89.9 ± 0.3	2.2 ± 0.2	14.6 ± 0.4	13.3 ± 0.4
36	89.0 ± 0.1	3.2 ± 0.0	12.6 ± 0.3	11.4 ± 0.2
39	90.1 ± 0.3	1.9 ± 0.0	12.1 ± 0.6	10.8 ± 0.6

Table 4.10: Results obtained for the optical properties of agar-agar powder in pouch made from pectin film at chill temperature.

Day	L*	a*	b*	Total Colour Difference (ΔE)
0	94.3 ± 0.0	1.4 ± 0.0	15.3 ± 0.0	15.0 ± 0.0
27	91.4 ± 2.2	1.5 ± 0.2	14.9 ± 1.4	13.9 ± 1.8
31	91.0 ± 0.0	1.7 ± 0.1	13.6 ± 0.1	12.5 ± 0.1
34	90.4 ± 0.0	1.3 ± 0.3	12.6 ± 0.7	11.3 ± 0.4
39	90.3 ± 0.1	1.5 ± 0.3	12.1 ± 0.2	10.8 ± 0.2

Table 4.11: Results obtained for the optical properties of agar-agar powder in pouch made from pectin papaya puree-based film at chill temperature.

Day	L*	a*	b*	Total Colour Difference (ΔE)
0	94.3 ± 0.0	1.4 ± 0.0	15.3 ± 0.0	15.0 ± 0.0
27	90.6 ± 1.4	1.4 ± 0.1	15.1 ± 1.1	13.9 ± 1.3
31	90.2 ± 0.4	1.6 ± 0.0	12.7 ± 0.1	11.4 ± 0.2
34	90.1 ± 1.0	1.5 ± 0.7	13.0 ± 0.5	11.7 ± 0.7
39	91.6 ± 1.0	1.3 ± 0.0	12.9 ± 0.9	11.9 ± 1.1

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS FOR FUTURE WORK

5.1 CONCLUSION

In conclusion, this project aimed which to explore the characterization and storage studies of soluble papaya puree pectin-based films on agar-agar powder has been achieved. The results showed that soluble pectin film has a higher solubility percentage than soluble papaya puree pectin-based film, with solubility percentages of 93.7% ± 5.8 and 84.5% ± 5.2 for soluble pectin film and papaya puree pectin-based film, respectively. Tensile strength (TS) was 4.8 ± 0.1 MPa, elongation at break (EAB) was 54.9% ± 3.7, Young Modulus (YM) was 0.17 ± 0.0, and toughness was 1.77 ± 0.02 MJ/m^3 for the soluble papaya puree pectin-based film. Meanwhile, the soluble pectin film had TS, EAB, YM, and toughness values of 4.3 ± 0.1 MPa, 42.1% ± 1.2, 0.19 ± 0.0, and 1.1 ± 0.0 MJ/m^3, which were all slightly lower than the soluble papaya puree pectin-based film.

The soluble papaya puree pectin-based film also had a higher seal strength than the soluble pectin film, with a seal strength of 0.5 ± 0.1 N/mm for the papaya puree pectin-based film and 0.4 ± 0.0 N/mm for the pectin film. Furthermore, the water vapour permeability (WVP) of soluble papaya puree pectin-based film was higher than that of soluble pectin film, with WVP values for papaya puree pectin-based film is 116.3^{-8} g /m. day Pa ± 33.2 and 115.5^{-8} g /m. day Pa ± 9.9 for pectin film. Furthermore, when compared to soluble pectin film, the optical properties of the soluble papaya puree pectin-based film revealed that the papaya puree pectin-based film was less transparent and opaque. The surface of soluble papaya puree pectin-based film is rougher than that of soluble pectin film, according to the AFM test.

The moisture content of agar-agar powder packed with soluble papaya puree pectin-based film film was lower than that of agar-agar powder packed with soluble pectin film at both storage temperatures in the studies of quality changes on the agar-agar powder. Meanwhile, on both storage temperatures and types of soluble film, the true density of agar-agar powder decreased. On both storage temperatures, the colour of agar-agar powder degraded. The decrease in total colour difference (ΔE) across all storage temperatures and film types indicates that the agar-agar powder loses its yellowish colour intensity over time. As a result, both objectives were met, and it is clear that the addition of papaya puree improved the pectin film's characteristics as well as reduce the quality changes in agar-agar powder during storage.

Soluble films were made using pectin, glycerol as a plasticizer, and fruit purees to serve as new soluble films in this study. Fruit purees were added to the film-forming solution to improve the mechanical properties. Due to its fibrous structure, papaya puree appears to be a promising candidate for incorporating pectin, as it improves the film’s mechanical properties. It also has a nice and appealing appearance and is easy to handle because it is not easily brittle. Moreover, fruit purees into pectin film also significantly improved the seal strength properties due to the interaction of film-forming material with the heat presence. Thus, it also shows potential in food packaging application. This innovative film packaging material is valuable as a useful alternative for food packaging applications. This pectin-based soluble film is not only low in preservatives and non-toxic in film-forming material, but it is also safe to eat.

5.2 RECOMMENDATIONS FOR FUTURE WORK

Some limitations are identified in the current study. Thus, in-depth studies of pectin films should be continued. The papaya puree used in this study showed a good seal strength value for the films. However, there is still much work on product storage study, such as water solubility test of the pouch with the product at various storage conditions. The test of the packaging testing should be done to determine the solubility of the product with the soluble packaging. Additionally, developing soluble film as a substitute for synthetic polymers can aid in testing films' active potential in a wider range of food packaging systems. It can also check the compatibility of natural films on food goods in terms of interaction with the matrix, in addition to optimizing the variable that would influence bio-packaging.

The shelf life or expiry of the films is one of the important criteria to film manufacturers. Other film's characterization like chemical migration or exposure to sunlight, can be performed on the soluble pectin and papaya puree-pectin based film. These pectin-based films can also be stored for a longer time to know when they start to fail the performance criteria be for use as packaging film. The suitable formulated pectin-based films can fulfill the market demand for environmentally film packaging, by knowing this information. Thus, this research may aid the food industry in achieving zero food packaging waste and may pose a positive impact against on environmental pollution.

REFERENCES

7 Types of Plastic that You Need to Know – Waste4Change. (n.d.). Retrieved July 25, 2021, from https://waste4change.com/blog/7-types-plastic-need-know/

Addala, R. (2015). Effect of Storage Conditions on Rate of Color Degradation of Paprika based Products. *Journal of Food Processing & Technology*, *06*(03). https://doi.org/10.4172/2157-7110.1000423

Ajay, S. M., Lalit, Y., Dash, M. B. S. K., & Mahanti, N. K. (2019). Application of Biodegradable Polymers in Food Packaging Industry : A Comprehensive Review. *Journal of Packaging Technology and Research*, *3*(1), 77–96. https://doi.org/10.1007/s41783-018-0049-y

Altaf, Uzma, Immanuel, Genitha, Iftikhar, & Farheena. (2018). *EXTRACTION AND CHARACTERIZATION OF PECTIN DERIVED FROM PAPAYA (Carica papaya Linn .) PEEL EXTRACTION AND CHARACTERIZATION OF PECTIN DERIVED FROM PAPAYA*. *3*(October), 1–6.

Alves, V. D., Castelló, R., Ferreira, A. R., Costa, N., Fonseca, I. M., & Coelhoso, I. M. (2011). Barrier properties of carrageenan/pectin biodegradable composite films. *Procedia Food Science*, *1*, 240–245. https://doi.org/10.1016/j.profoo.2011.09.038

Arham, R., Mulyati, M. T., Metusalach, M., & Salengke, S. (2016). Physical and mechanical properties of agar based edible film with glycerol plasticizer. *International Food Research Journal*, *23*(4), 1669–1675. https://doi.org/10.31227/osf.io/tq2pf

Article, R. (2011). Basketful Benefits of Papaya. *International Research Journal of Pharmacy*, *2*(7), 6–12.

BeMiller, J. N. (1986). *An Introduction to Pectins: Structure and Properties*. 2–12. https://doi.org/10.1021/bk-1986-0310.ch001

Cho, S. Y., Lee, S. Y., & Rhee, C. (2010). Edible oxygen barrier bilayer film pouches from corn zein and soy protein isolate for olive oil packaging. *LWT - Food Science and Technology*, *43*(8), 1234–1239. https://doi.org/10.1016/j.lwt.2010.03.014

Eça, K. S., Machado, M. T. C., Hubinger, M. D., & Menegalli, F. C. (2015). Development of Active Films From Pectin and Fruit Extracts: Light Protection, Antioxidant Capacity, and Compounds Stability. *Journal of Food Science*, *80*(11), C2389–C2396. https://doi.org/10.1111/1750-3841.13074

Go, E.-J., & Song, K. Bin. (n.d.). *Development and Characterization of Citrus Junos Pomace Pectin Films Incorporated With Rambutan (Nephelium Lappaceum) Peel Extract*. https://doi.org/10.3390/coatings10080714

Grigore, M. E. (2017). Methods of recycling, properties and applications of recycled thermoplastic polymers. *Recycling*, *2*(4), 1–11. https://doi.org/10.3390/recycling2040024

Kwok, D. C. Y., & Liang, D. D. S. S. (2019). *Biology of papaya*. http://www.biosafety.gov.my/ms-my/pustaka-media/Documents/biological document-papaya.pdf

Langer, J. (1969). Chemical Properties and Derivatives of Glycerol. *Annals of Physics*. http://www.mendeley.com/research/no-title-avail/

Liu, C., Huang, J., Zheng, X., Liu, S., Lu, K., Tang, K., & Liu, J. (2020). Heat sealable soluble soybean polysaccharide/gelatin blend edible films for food packaging applications. *Food Packaging and Shelf Life*, *24*(November 2019), 100485. https://doi.org/10.1016/j.fpsl.2020.100485

Maryam Adilah, Z. A., Jamilah, B., & Nur Hanani, Z. A. (2018). Functional and antioxidant properties of protein-based films incorporated with mango kernel extract for active packaging. *Food Hydrocolloids*, *74*, 207–218. https://doi.org/10.1016/j.foodhyd.2017.08.017

Nazmi, N. N. M., Isa, M. I. N., & Sarbon, N. M. (2020). Characterisation of Biodegradable Protein Based Films from Gelatin Alternative: a Review. *International Food Research Journal*, *27*(6), 971–987.

Nur, S., Binti, H., & Raduan, M. (2020). *TENSILE PROPERTIES , HEAT SEALABILITY AND PEEL STRENGTH OF PECTIN-BASED EDIBLE FILM AND PACKAGE PROJECT REPORT SUBMITTED IN PARTIAL FULFILLMENT OF THE*. 1–66.

Otoni, C. G., Avena-Bustillos, R. J., Azeredo, H. M. C., Lorevice, M. V., Moura, M. R., Mattoso, L. H. C., & McHugh, T. H. (2017). Recent Advances on Edible Films Based on Fruits and Vegetables—A Review. *Comprehensive Reviews in Food Science and Food Safety*, *16*(5), 1151–1169. https://doi.org/10.1111/1541-4337.12281

Pathare, P. B., Opara, U. L., & Al-Said, F. A. J. (2013). Colour Measurement and Analysis in Fresh and Processed Foods: A Review. *Food and Bioprocess Technology*, *6*(1), 36–60. https://doi.org/10.1007/s11947-012-0867-9

Paull, R. E., Nishijima, W., Reyes, M., & Cavaletto, C. (1997). Postharvest handling and losses during marketing of papaya (Carica papaya L.). *Postharvest Biology and Technology*, *11*(3), 165–179. https://doi.org/10.1016/S0925-5214(97)00028-8

Pawar, P. a, & Purwar, A. H. (2013). Bioderadable Polymers in Food Packaging. *American Journal of Engineering Research (AJER)*, *05*, 151–164.

Sanyang, M. L., Sapuan, S. M., Jawaid, M., Ishak, M. R., & Sahari, J. (2015). Effect of plasticizer type and concentration on tensile, thermal and barrier properties of biodegradable films based on sugar palm (Arenga pinnata) starch. *Polymers*, *7*(6), 1106–1124. https://doi.org/10.3390/polym7061106

Sherrington, D. C. (1992). Functional polymers and their applications. *Reactive Polymers*, *18*(1), 87. https://doi.org/10.1016/0923-1137(92)90297-f

Sila, D. N., Van Buggenhout, S., Duvetter, T., Fraeye, I., De Roeck, A., Van Loey, A., & Hendrickx, M. (2009). Pectins in processed fruits and vegetables: Part II-Structure-function relationships. *Comprehensive Reviews in Food Science and Food Safety*, *8*, 86–104.

Sothornvit, R., & Pitak, N. (2007). Oxygen permeability and mechanical properties of banana films. *Food Research International*, *40*(3), 365–370. https://doi.org/10.1016/j.foodres.2006.10.010

Souza, A. C., Benze, R., Ferrão, E. S., Ditchfield, C., Coelho, A. C. V., & Tadini, C. C. (2012). Cassava starch biodegradable films: Influence of glycerol and clay nanoparticles content on tensile and barrier properties and glass transition temperature. *LWT - Food Science and Technology*, *46*(1), 110–117. https://doi.org/10.1016/j.lwt.2011.10.018

Suput, D., Lazic, V., Popovic, S., & Hromis, N. (2015). Edible films and coatings: Sources, properties and application. *Food and Feed Research*, *42*(1), 11–22. https://doi.org/10.5937/ffr1501011s

Thakur, B. R., Singh, R. K., & Handa, A. K. (1997). Chemistry and Uses of Pectin - A Review. *Critical Reviews in Food Science and Nutrition*, *37*(1), 47–73. https://doi.org/10.1080/10408399709527767

Tharanathan, R. N. (2003). Biodegradable films and composite coatings: Past, present and future. *Trends in Food Science and Technology*, *14*(3), 71–78. https://doi.org/10.1016/S0924-2244(02)00280-7

The time is now for edible packaging. (n.d.). Retrieved July 25, 2021, from https://cen.acs.org/food/food-science/time-edible-packaging/98/i4

Tulamandi, S., Rangarajan, V., Rizvi, S. S. H., Singhal, R. S., Chattopadhyay, S. K., & Saha, N. C. (2016). A biodegradable and edible packaging film based on papaya puree, gelatin, and defatted soy protein. *Food Packaging and Shelf Life*, *10*, 60–71. https://doi.org/10.1016/j.fpsl.2016.10.007

What are the benefits of biodegradable water soluble plastic film ? (n.d.). Retrieved August 10, 2021, from https://www.soltec-development.com/en/benefits-biodegradable-water-soluble-plastic-film/

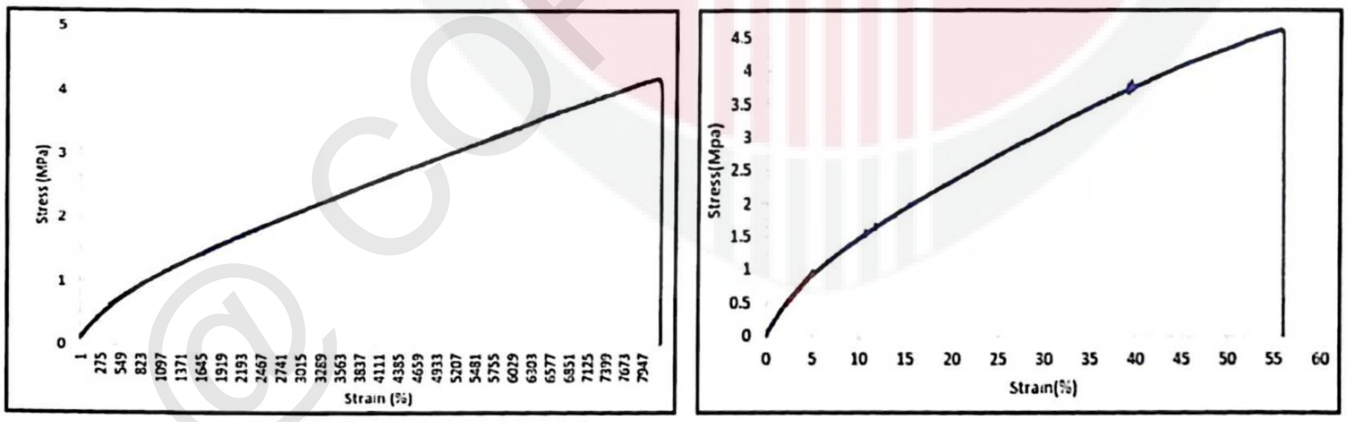
Yai, H. (2008). Edible films and coatings : characteristics and properties. *International Food Research Journal*, *15*(3), 237–248.

Yu, A. B., Standish, N., & Lu, L. (1995). Coal agglomeration and its effect on bulk density. *Powder Technology*, *82*(2), 177–189. https://doi.org/10.1016/0032-5910(94)02912-8

APPENDIX

Appendix 1: Tensile properties values and graph of stress and strain of pectin film and papaya puree pectin-based films.

Test ID	Tensile Strength (MPa)	EAB %	Young Modulus (stress/strain) Energy per unit volume to break	Toughness MJ/m^3
(Thickness = 0.084 mm)				
Pectin film 1	4.190	41.072	0.193	1.025
Pectin film 2	4.357	43.382	0.172	1.107
Pectin film 3	4.335	41.912	0.196	1.065
Average:	4.29	42.122	0.187	1.07
S.D.	0.09	1.17	0.01	0.04
(Thickness= 0.10568mm)				
PP - 1	4.6331	56.046	0.141	1.57
PP - 2	5.169	57.808	0.166	1.82
PP - 3	4.5218	50.732	0.211	1.91
Average:	4.77	54.862	0.173	1.77
S.D.	0.35	3.684	0.035	0.18



Graph of stress and strain of pectin film and papaya puree pectin-based films.

Appendix 2: Result for optical for pectin film and pectin papaya puree-based film.

Pectin papaya puree film - PP thickness = 0.10568mm

sample	L*	a*	b*	T600(%)	A600	opacity	transparency	delta E
PP1	88.00	2.42	19.83	81.060	0.091	0.863	0.863	23.77
PP2	87.99	2.38	20.13	81.130	0.091	0.859	0.859	24.02
PP3	88.53	2.14	19.16	82.000	0.086	0.816	0.816	22.90
PP4	88.31	2.04	19.04	80.980	0.092	0.867	0.867	22.91
PP5	88.51	2.00	18.93	81.390	0.089	0.846	0.846	22.70
average	88.27	2.20	19.42	81.312	0.090	0.850	0.850	23.26
SD	0.26	0.19	0.53	0.414	0.002	0.021	0.021	0.59

Pectin film thickness = 0.084mm

sample	L*	a*	b*	T600(%)	A600	opacity	transparency	delta E
control1	92.11	-0.97	9.61	84.11	0.0752	0.8947	0.8947	13.0637
control2	92.16	-0.82	8.95	83.89	0.0763	0.9082	0.9082	12.5415
control3	91.89	-0.86	9.52	83.50	0.0783	0.9323	0.9323	13.1402
control4	92.05	-0.93	9.42	83.82	0.0767	0.9125	0.9125	12.9630
control5	91.50	-0.85	9.50	82.68	0.0826	0.9833	0.9833	13.3968
average	91.94	-0.89	9.40	83.60	0.0778	0.9262	0.9262	13.0210
SD	0.27	0.06	0.26	0.56	0.0029	0.0347	0.0347	0.3125

Appendix 3: Data for moisture content of agar-agar powder on pouch made from pectin film and papaya puree pectin-based film.

pectin film ambient temperature.

Time(days)	A (%)	B (%)	average moisture (%)
0	11.8	11.8	11.8± 0.0
7	20.5	20.5	20.5± 0.0
32	20.2	18.9	19.51± 0.9
36	19.5	20.5	20.0± 0.7
39	20.5	20.0	20.2± 0.4

pectin film chill temperature.

Time(days)	A (%)	B (%)	average moisture (%)
0	11.8	11.8	11.8 ± 0.0
27	21.2	20.5	20.9 ±0.5
31	20.6	20.2	20.4 ±0.4
34	20.5	20.5	20.5±0.0
39	20.6	20.0	20.4±0.3

papaya puree pectin-based film ambient temperature.

Time(days)	A (%)	B (%)	average moisture (%)
0	11.8	11.8	11.8± 0.0
7	19.8	20.0	19.9± 0.1
32	18.9	19.4	19.1± 0.4
36	18.9	19.4	19.2± 0.4
39	19.3	19.5	19.4± 0.1

papaya puree pectin -based film chill temperature

Time(days)	A (%)	B (%)	average moisture (%)
0	11.8	11.8	11.8± 0.0
27	20.4	19.6	20.0± 0.6
31	20.3	20.3	20.3± 0.0
34	20.0	20.2	20.1± 0.2
39	20.1	20.0	20.1± 0.0

Appendix 4: Data for the moisture content of agar-agar powder during storage in pouch made from pectin film and papaya puree pectin -based film.

pectin film ambient temperature.		
Time(days)	Average vol. (cm^3)	average density (g/cm^3)
0	0.91± 0.00	1.41± 0.00
7	0.86± 0.01	1.36 0.00
32	0.81± 0.11	1.36± 0.02
36	0.77 ± 0.04	1.36± 0.07
39	0.78± 0.10	1.36± 0.02

pectin film chiller temperature.		
Time(days)	Average vol. (cm^3)	average density (g/cm^3)
0	0.91± 0.00	1.41 ± 0.00
27	0.75± 0.00	1.36± 0.00
31	0.78 ± 0.02	1.36± 0.03
34	0.81± 0.00	1.36± 0.07
39	0.81± 0.12	1.36± 0.02

papaya puree pectin -based film ambient temperature.		
Time(days)	Average vol. (cm^3)	average density (g/cm^3)
0	0.91 ± 0.00	1.41± 0.00
7	0.75± 0.00	1.36± 0.00
32	0.76± 0.05	1.36± 0.03
36	0.80± 0.07	1.36± 0.07
39	0.79± 0.00	1.36± 0.02

papaya puree pectin -based film chiller temperature.		
Time(days)	Average vol. (cm^3)	average density (g/cm^3)
0	0.91± 0.00	1.41 ± 0.00
7	0.88± 0.10	1.36± 0.00
32	0.77± 0.01	1.36± 0.03
36	0.81± 0.05	1.36± 0.07
39	0.89± 0.00	1.36± 0.02