Characterization of Edible Film Made of Pectin from Nutmeg and Palmitic Acid

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Abstract: The development of biopolymers as a packaging material is increasingly needed to reduce environmental pollution due to the use of synthetic plastics which are biologically difficult to break down. Nutmeg contains pectin which can be used as a base for making edible films. The aim of the study was to determine the characterization of the edible film from the pectin of nutmeg with the addition of palmitic acid and glycerol as a plasticizer. The results showed that the increase in the concentration of pectin and palmitic acid tended to significantly increase the thickness of the edible film, the elongation rate and the tensile strength of the film, but it could decrease the water vapor transmission rate and the film solubility. The best treatment was a concentration of 30% wet pectin (w / v) and palmitic acid 0.04% (w / w) which resulted in the lowest water vapor transmission rate of 2.39 (g / mm / m^2 hour).

1 INTRODUCTION

Packaging is one way to protect food from the influence of internal and external factors that cause damage. Thus the role of packaging materials is very important.*Plastic* is the most widely *used* for *packaging* material, *but plastic have weaknesses such as non-biodegradable* properties, which remain in the environment for long periods of time, they remain threats to the environment (Krochta and Johnston 1997).

The need for packaging materials for developed countries reaches 250 kg / per capita / year, while for countries that are starting to develop and are poor, is 5 kg / capita / year. Approximately 30% of the total residential solid waste is packaging material and 13% of this amount is plastic waste. In other words, plastic waste reaches 4% of the total residential solid waste. The biggest demand for packaging is in the form of flexible packaging which reaches 42% of total sales. Followed by Paper Board Packaging 28%, Rigid Plastic Packaging 14%, Woven Polyolefin Sack 6%, Metal Can Packaging 5%, Glass Container Packaging 3% and others 2%. Due to the nature of plastic which is difficult to break down naturally, its existence starts to pollute the environment. However, plastic has the advantages of being lightweight, strong and economical. With these superior properties, it is estimated that if the plastic is not replaced with other packaging materials, it is estimated that the weight of packaging waste will increase by 100%, the volume will increase by 250% and the cost will increase by 250%. Therefore, it is necessary to develop other packaging materials that have superior properties such as plastic which is strong, lightweight and economical as well as being biodegradable and even edible.

In developed countries such as America, it has responded to this challenge by developing edible films using organic biopolymers from agricultural products (Dumat, 1999). Edible film is a thin material that covers a food ingredient and is safe for consumption. Edible film serves as a barrier against the transfer of water vapor, aroma, oxygen and gases as well as protecting against microbial attack. Usually, the function of edible films is enhanced by adding antioxidants, antimicrobials, nutrients and other food additives. The basic ingredients for making edible films are biopolymers of agricultural products including waste products such as protein (corn, milk, wheat, soybeans) while carbohydrates (starch, pectin, alginate) and fat. The use of carbohydrates as a basic material for making edible films is based on relatively cheap costs, abundance of materials, and thermoplastic properties (Renata et al.,

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2014). One of the sources of carbohydrates is pectin. The results of the research by Layuk, 2001 reported that the old nutmeg contains 21.54% pectin so that it has the potential to be used as a base for making edible films. The edible film made from nutmeg pectin does not experience damage (moldy) after being stored at room temperature due to the presence of polyphenols in the nutmeg which can inhibit the growth of fungi on the edible film. Edible films made from carbohydrates such as pectin, starch and alginate have disadvantages, including easy hydration, expands quickly and tears easily. To overcome this, it is necessary to add fatty acids (McHugh and Krochta 1994). Edible film requires a plasticizer. Plasticizer can flex and prevent the brittleness of the edible film. Glycerol is a plasticizer, which is commonly used in making edible films (Han, 2005). Glycerol contains relatively small hydrophilic molecules and easily inserted between the polymer chains of the base material. This condition causes structural modification of the molecules making up the edible film. Glycerol molecules will disrupt the polymer cohesiveness of the base material by reducing intermolecular interactions and increasing polymer mobility thereby improving the flexibility and extensibility of the edible film.

Permeability concerns the process of transferring solution and diffusion when the solution moves from one side of the film and then diffuses to the other side of the film. The thicker the edible film produced, the better the ability of the edible film to hold water vapor. Fatty acids such as glycerol and palmitic acid have hydrophilic groups that reduce molecular density so that they can form free space in the film matrix which facilitates diffusion of water vapor (Ruan et al. 1998). Several studies have combined two types of materials to improve the quality of edible films such as; breadfruit and chitosan (Setiani et al., 2013), whey protein concentrate and / or with mesquito gum / sodium alginate / caragenate (Villagomez-Zavala, et al., 2008), pectin and tapioca (Layuk, 2001), sago starch and carrageenan (Anggraini 2012). Nutmeg pectin and sago starch (Layuk, et al, 2019), lindur fruit starch with carrageenan (Jacoeb, 2014), soybean and tapioca extract (Sinaga et al, 2013), Dangke and Agar (Fatma, et al, 2015), starch sweet potato with glycerol (2018) and nutmeg pectin and sago starch (Layuk, et al, 2019).

The research aimed to develop nutmeg pectin as a raw material for making edible films with the addition of plasticizers (palmitic acid) in various concentrations and their effects on the characteristics of the films produced.

2 MATERIALS AND METHODS

2.1 Materials and Tools

The materials used in the study were nutmeg, palmitic acid, glycerol, calcium chloride (CaCl2), distilled water, HCl and alcohol. The tools used is a digital scale, beaker glass, measuring cup, hot plate stirrer, magnetic stirrer, thermometer, glass plate measuring 8 x 7 x 2 cm, oen, Ioyid Instrument, micrometer and other auxiliary equipment.

2.2 Research Methods

The research was carried out at the Laboratory of the North Sulawesi Agricultural Technology Study Center and the Laboratory of the Faculty of Agricultural Technology, UGM Yogyakarta, from March to December 2019. The stages of the research included the isolation of pectin, making edible film and the characteristics of the edible film.

2.3 Pectin Isolation (Layuk 2001)

Nutmeg pectin is obtained through several stages. Ripe nutmeg is washed and cut in half to separate the seeds and pulp. The pulp of the nutmeg is then cut into 2 x 2 cm cubes. Then blanched for 5 minutes in boiling water to activate the enzyme. Then dried in an oven at 50 C for 8-12 hours until the moisture content reaches 10-11%. Dried nutmeg is ground with a fineness level of 50 mesh. Then the nutmeg powder was extracted using HCl pH 2.0 at a temperature of 80 C for 2 hours. The schematic of pectin extraction and isolation is shown in Figure 1.

2.4 Edible Film Making

Nutmeg pectin with concentrations of 10% (A1), 20% (A2), 30% (A3) and 40% (A4) w / v and palmitic acid 0% (B0), 0.02% (B1), 0, 04% (B2) and 0.06% w / w pectin. The solution is made by adding pectin to 100 ml of distilled water which already contains 2% sago starch. , then heated at a temperature of 85 C for 15 minutes while stirring with a magnetic stirrer. After that the temperature was lowered to 40 C, then added palmitic acid and glycerol. The heating was continued again until the temperature was 85 C while stirring with magnetic stirrer for 10 minutes until the solution was homogeneous. The solution is then poured into a glass plate mold measuring 8 x 7 x 2 cm (length x width x thickness), dried at 50 C for 10-12 hours (Figure 2). The film was then removed from the mold and stored in a plastic container filled with silica gel

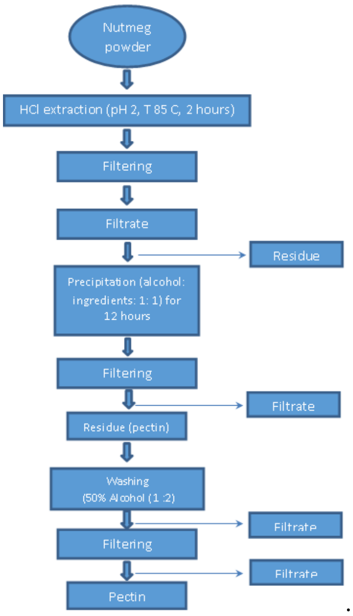

Figure 1: Pectin isolation.

for 12 hours. Relative humidity in a plastic container is around 40-50%. The film is then cut into pieces according to the parameters to be tested. Tensile strength and elongation tests were 3 x10cm, for WVTR a circle was made with a diameter of 7 cm. The solubility is 2 cm x 2 cm. Before taking measurements, the film was conditioned for 24 hours in a plastic container containing silica gel.

The experiment used was factorial completely randomized with three replications (Gomez and Gomez, 1995)

2.5 Data Analysis and Collection Procedure

Measurement of water content, ash content and protein content (AOAC, 1984), methoxyl and polygalaruronic concentrations (Rangana, 1977). Testing thickness, tensile strength and elongation (Gontard, et al, 1992) tensile strength and elongation were measured using a Universal Testing Machine (LIoyd Instrument), while film thickness was measured using a micrometer (accuracy 0.001 mm).

2.6 Film Solubility (Gontard et al, 1992)

The percentage of film solubility was determined by heating the film at 100 C for 24 hours with two pieces of edible film with a size of 2 x 2 cm. Weighed then immersed in 50 ml of water containing 0.02% sodium azide. The immersed edible film was stored at 20 C for 24 hours. Then the film was taken and dried at 100 C for 24 hours. To determine the solubility of the edible film, it was calculated by subtracting the initial weight minus the insoluble dry weight multiplied by 100%.

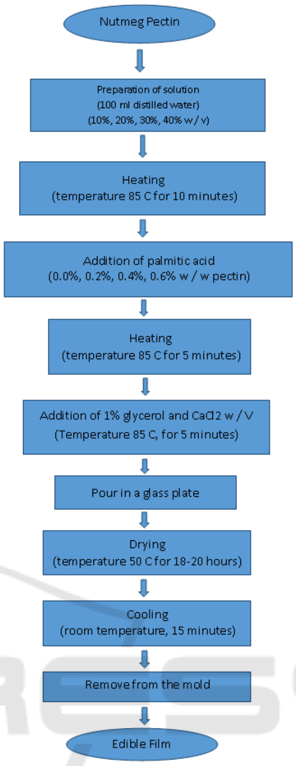

Figure 2: Edible film making.

2.7 Water Vapor Transmission Rate WVTR Testing (ASTM, 1980)

The water vapor transmission rate (WVTR) using the gravimetric method was determined using the ASTM procedure (1980). The film to be tested is glued to a bowl made of acrylic with an outer diameter of 8 cm, an inner diameter of 7 cm and a thickness of 2 cm, then filled with 50 ml of water. The acrylic bowl is then stored in a jar containing silica gel. Relative humidity 20%. The water vapor that diffuses through the film will be absorbed by the silica gel, the amount of which can be found by calculating the reduced weight of the bowl filled with water at the time of measurement. Weighing was done at intervals, 1,2,3,4,5,6 and 7 hours. All tests were carried out twice. Changes in bowl weight and time are then plotted on a graph where the y-axis is the weight of the plate (g) when weighed and the x-axis is time (from this graph a regression line equation will be generated where the resulting slope is the rate of weight gain per unit (g / hour). WVTR is calculated by dividing the slope by the area of the film, so that the WVTR unit is g / m2h.

3 RESULTS AND DISCUSSION

3.1 Nutmeg Pectin Identification

The results of the composition analysis and identification of pectin are as shown in Table 1. The results showed that the pectin obtained was high methoxyl pectin. Towle and Chistinsesn (1973) stated that pectin which has a methoxyl count of 7-14% is a high methoxyl pectin. Meanwhile, if the methoxyl content is 0 -7%, it is a low methoxyl pectin. The galacturonic content obtained was 84.18% and compared to the commercial pectin the level was 83.78%, it can be said that the pectin produced has a level of purity. This is supported by the low ash content of 1.49%. High methoxyl and polygalacturonic content and low ash content indicate the level of purity of pectin and the factors that play a role in the gel formation of a pectin. The presence of protein content can also support the success of nutmeg pectin as a basic material for making edible films.

Table 1: Nutmeg pectin composition with commercial pectin.

Composition	Nutmeg pectin *	Commercial pectin **)
Yield (%)	21,54	-
Water content (%)	8,95	11,05
Ash content (%)	1,49	3,42
Protein Content (%)	4,37	1,95
Methoxyl content (%)	11,48	11,42
Polygalacturunoate content (%)	84.17	83.78

3.2 Film Thickness

The thickness of the films obtained ranged from 0.03 to 0.07 mm. The highest thickness was obtained at 3% pectin concentration and 0.6% palmitic acid (A3B3). In Figure 3, it can be seen that the higher the concentration of pectin and palmitic acid, the thicker the film produced. This occurs because the increasing number of film-forming components, resulting in the thicker film produced. The thickness of the film is influenced by the amount of solids in the film forming solution and the thickness of the mold. With the addition of glycerol as a plasticizer, it can reduce the thickness and force density between pectin molecules so that the chain movement is good and the resulting film is more flexible and smoother. This is possible because glycerol is a small hydrophilic molecule that can easily enter between pectin chains and form bonds (Gontard et al, 1992, Layuk et al, 2019). The same thing was reported by Sinaga et al, (2013) that the addition of plasticizers such as glycerol has an effect on the thickness of the edible film, elongation and tensile strength of the film.

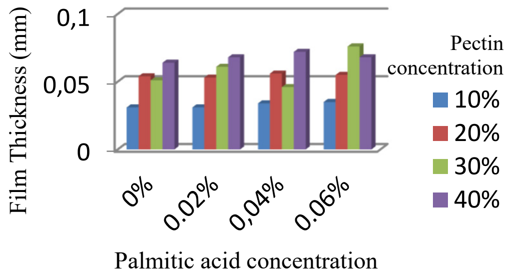

Figure 3: Edible film thickness at various concentrations of pectin and palmitic acid.

3.3 Elongation

Film elongation is a mechanical property related to the chemical structure of the film. Film elongation is the change in the maximum elongation experienced by the film during the tensile strength test at the time the film is torn (Hay, 1968). According to Gontard and Gilbert (1993), the mechanical properties of films depend on the type of material, especially the structural cohesion properties. This property is a result of the polymer's ability to form strong molecular bonds. The results of variance can be seen that there is a very significant effect between the pectin concentration on the percentage of film elongation. The palmitic acid concentration of 0.2% and without the addition of palmitic acid had a high elongation compared to other treatments, both at 10%, 20% and 30% pectin concentrations. This is because palmitic acid has anti-plasticizer properties against pectin edible film. Gontard and Gulbert (1993) said that fatty acids have anti-plasticizing properties in films made from gluten. The same thing was reported (Layuk, 2001 and Layuk et al, 2019) that the anti-plasticizing properties of palmitic acid in pectin were caused by the formation of a galacturonic-fatty acid complex which will add to the three-dimensional network integrity of the polymer film, thereby reducing the elongation rate of the film. Meanwhile, Min Lai and Hucy (1997) reported that palmitic acid can act as a plasticizer that can change the mechanical characteristics of edible films. The addition of palmitic acid will be able to increase the tensile strength of the film breaking. When it reaches a critical point, the addition of palmitic acid (Park et al., 2004) and glycerol (Fantasari et al) will decrease the elongation and tensile strength of the film.

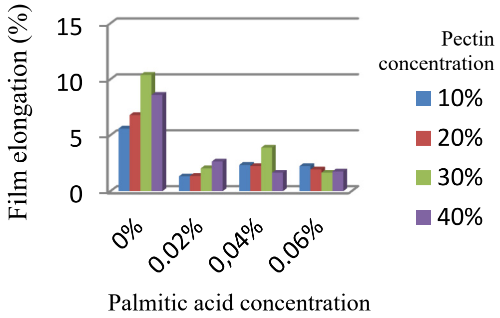

Figure 4: Edible film extension at various concentrations of pectin and palmitic acid.

3.4 Tensile Strength

Tensile Strength is a mechanical property related to the chemical structure of the film. Tensile strength is defined as the maximum tensile stress that the sample can accept. The mechanical properties of the edible film depend on the film-forming material, especially the structural cohesion properties. This property is a result of the polymer's ability to form strong molecular bonds Gontard and Gulbert (1993). In Figure 5, it can be seen that the higher the concentration of pectin and palmitic acid the higher the resulting tensile strength. The highest tensile strength occurred in the treatment with pectin concentration of 30% and palmitic acid 0.6%, which is 8.71 kPa. This is because the galacturonic-palmitic acid complex can add to the polymer integration of the edible film so that the resulting tensile strength is higher. The addition of palmitic acid and glycerol can improve the mechanical properties of the film where the film is stronger, more compact and flexible. The addition of palmitic acid also increases the number of carbons and functional groups in the matrix chain thereby increasing the tensile strength and elongation (Donhowe and Fennema 1994).

3.5 Edible Film Solubility

Solubility in a solution is one of the determining factors in choosing edible film as a packaging material. Figure 6 shows that the solubility of the film decreases as more palmitic acid is added. This is due to palmitic acid which has a high C chain (C16) making it difficult to dissolve in water. According to Koelsch and Labuza (1992), palmitic acid can dissolve in polar and non-polar organic solvents. The ability to

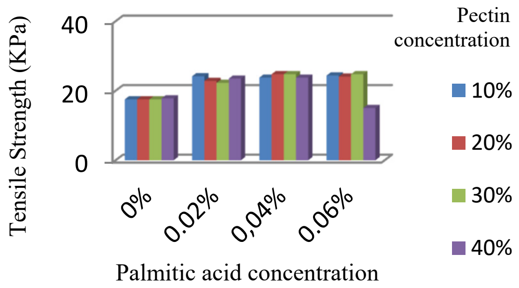

Figure 5: Tensile strength of edible films at various concentrations of pectin and palmitic acid.

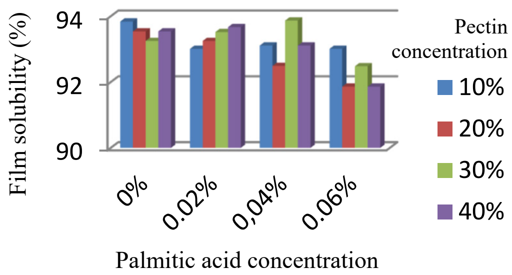

Figure 6: Edible film solubility at various concentrations of pectin and palmitic acid.

dissolve in water is influenced by the C chain. The longer the C chain of fatty acids, the more difficult their solubility in water is.

3.6 Water Vapor Transmission Rate of Film (WVTR)

The ability of the film to withstand the water vapor transmission rate can be seen in Figure 7. The resulting water vapor transmission rate ranges from 2.39 g / mm / m2 / hour - 5.10 g / mm / m2 / hour. The highest WVTR was obtained at treatment concentrations of pectin 40% and palmitic acid 006%. The high WVTR is due to the increasing number of hydrophilic components in the film which causes water absorption on the film and increases the weight of the film during the WVTR measurement. This proves that biodegradable films require hydrophobic materials to reduce the rate of water vapor transmission through the film. While the lowest was at the pectin concentration of 30% and palmitic acid 0.4%. According to Hui (2006), palmitic acid can act as a plasticizer that can change the mechanical characteristics of edible film, which can increase hydrogen bonding thereby reducing water vapor transmission rates. However, when it reaches a critical point the addition of palmitic acid will reduce hydrogen bonding and increase the intermolecular space which will increase the flexibility of the film and provide a cavity that allows diffusion of penetrant molecules (Layuk 2001 and Slade and Levine, 1991). This is also due to the presence of glycerol which causes the speed of water vapor and gas transmission through the film to increase (Gontar et al., 1983) and due to reduced molecular density so that free space is formed in the film matrix so as to facilitate the diffusion of water and gas. The same thing was found in this study where the palmitic acid concentration of 0.6% both at 10%, 20% and 30% pectin concentration, resulted in increased WVTR and the resulting film surface was rather rough.

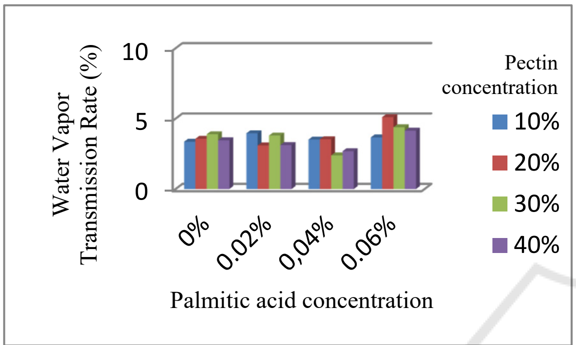

Figure 7: Water vapor Transmission Rate Edible film at various concentrations of pectin and palmitic acid.

4 CONCLUSIONS

- Nutmeg pectin can be used as material for making edible films.
- Increasing the concentration of pectin and palmitic acid can increase the thickness, elongation and tensile strength of the film, but can reduce WVTR and film solubility.
- The best treatment was pectin concentration of 30% and palmitic acid 0.4% where the smallest water vapor transmission rate was 2.39g / mm / m2 / hour.

REFERENCES

AOAC. 1984. Official Method of Analysis of Association of Official Analytical Chemists, 14thEd. AOAC Inc., Arlington,Virginia.

ASTM. 1980. *Standart Test Methods for Water Vapor Transmission Of Materials*. ASTM Book of Standards

Anggraini 2012. *Characteristics of Edible Film from Sago Starch with the Addition of Glycerol and Carrageenan*. Journal of Food and Agroindustry 1: 12-1

Bourtoom T. 2008. *Review article edible films and coatings: characteristics and properties*. Journal International Food Research 15(3):237-248.

Donhowe,G. and O. Fennema. 1994. *Edible film and Coting: Characteristics Formation, Definitions and Testing Methods: Krochta*, J.M., E.A.

Baldwin, and M.O.Nisperos-Carriedo (eds). Edible Coatings and Films to Improve Food Quality. Tecnomic Publ.Co.,Inc.,Lancaster, USA.

Dumat, 1996. *Making edible films from a mixture of rubber seed protein with casein*. Postgraduate Thesis.Master Program in Food and Science Technology. Faculty of Agricultural Technology UGM.

Fatnasari, A. K. A. Nocianitri and I Putu Suparthana. *The Effect of Glycerol Concentration on The Characteristic Edible Film Sweet Potato Starch (Ipomoea batatas L.)*. Scientific Journal Of Food Technology Vol. 5, No.1, 27 - 35, March 2018.

Fatma, Ratmawati Malaka and Muhammad Taufik.2015. *Characteristics of Edible Film Made from Dangle Whey and Agar Using Different Percentage of Glycerol*. JITP Vol. 4 No. 2, Page 63-69, July 2015

Fennema O, Donhowe IG, Kester JJ. 1994. *Lipid type and location of the relative humidity gradient influence on the barrier properties of lipid to water vapor*. Journal of Food Engineering 22(1):225-239

Gontar, N. Guilbert and J.L. Cug. 1992. *Water and Glycerol as Plasticizer affect mechanical and water vapor barrier properties of edible film wheat gluten film. J.* Food Sci: 58(1): 206-210

Gontard, N. and Gulbert,S. 1993. *Bio Packaging. Technology and properties of edible film and biodegradable material of agricultural origin*. In M. Methiouthi. Food packaging and preservation. The AVI Publ.Inc. Westport Connecticut.

Han, 2005. *Innovations in Food Packaging*. Department of Food Science University of Manitoba Wiminpeg, Manitoba Canada.

Hay,P.M. 1968. *Properties and Methods of Identification of Commercial Films*. In *O.J. (ed) .The Science and Technology of Polymer Film*, London.

Hui, Y. H. 2006. *Handbook of Food Science, Technology, And, Engineering Volume I*. Crc Press. USA.

Jacoeb M. A. Roni Nugraha, Siluh Putu Sri Dia Utari. 2014. *Edible Film from Lindur Fruit Starch with Addition of Glycerol and Carrageenan. JPHPI 2014*, Volume 17 No 1.

Koelsch C.M. and Labuza, T.P. (1992). *Functional physical and morphological properties of methyl cellulose and fatty acid-based edible barriers*. Lebensm Wiss U-Technol 25:404-411.

Krochta JM and Johnston CDM. 1997. *Edible and Biodegradable Polymer Film. Journal of Food Technology* 52(2): 1-20.

Layuk, P. 2001. *Characterization of Nutmeg Pectin Composite Edible Film with Tapioca.* PostgraduateThesis.Master Program in Food and Science Technology. Faculty of Agricultural Technology UGM

Layuk P . Sondakh J. Marietje P. *Characteristics and Permeability Properties of Sago Starch Edible Film. AGRITEKNO, Journal of Agricultural Technology Online Version:* http://ojs3.unpatti.ac.id/index.php/

agritekno Vol. 8, No. 2, 34-41, Th. 2019 DOI: 10.30598/jagritekno.2019.8.2.34 ISSN 2302-9218

McHugh TH, Krochta JM. 1994. *Sorbitol vs Glycerol plasticized whey protein edible films: integrated oxygen permeability and tensile strength property evaluation*. Journal of Agricultural and Food Chemistry 42(4):841-845.

Min Lai an Huey 1997. *Properties of microstructure of sheets plasticized with palmitic acid*. J. Cereal Chemiestry 42(2).

Park, D.P., Sung, J.H., Choi, H.J. and Jhon, M.S. (2004). *Electroresponsive characteristics of highly substituted phosphate starch*. Journal of Material Science 39: 60836086.

Park JW, Testin, RF, Vergano DJ, Park KJ, Weller CL, 1996. *Application of Laminated Edible Film to Potato Chip Packaging*. Journal of Food Science 61(4): 66-76

Rangana S. 1977. *Manual of Analysis of Fruit and Vegetable Product*. Tat MC. Grow Hill Publishing Company Limited New Delhi.

Renata C., dos Reis., I. A. Devilla, G. H. H. Oliveira, P. C. Corrêa, D. P. R. Ascheri, A. B. M. Souzay and A. C. O. Servulo. 2014. *Mechanical Properties, Permeability and Solubility Of Films Composed of Yam Starch and Glycerol*. INVERCIENCIA, 39: 410415.

Ruan RR, Xu L, Chen PL. 1998. *Water Vapor Permeability and Tensile Strength of Cellulose-Based Composite Edible Film Applied Engineering In Agriculture* 14(4):411-413.

Setiani, W., T. Sudiarti, L. Rahmidar. 2013. *Preparation and characteristics of edible films from polyglend breadfruit starch-chitosan*. Valensi, 3(2): 100-10.

Sinaga L.L., Melisa S. R .S, Mersi S. S. 2013. *Characteristics of Edible Film From Soybean Extract With The Addition Of Tapioca Flour And Glycerol As Food Packaging Materials USU's Chemical Engineering Journal*, Vol. 2, No. 4 (2013)

Slade and Levine, 1991. *Beyond water activity: Recent advances on an alternative approach to the assessment of food quality and safety*. Food Sec. 61(1): 192-194.

Towle, G.G. and Chistinsen, O. 1973. *Pectin, in R.L.Wister Industrial GUM*. Academic press New York.

Villagomez-Zavala, D. L., C. Gomez-Corona, E. S. M.Matinez and J. P. P. Orozco. 2008. *Comparative study the mechanical properties of edible films made from single and blended hydrophilic biopolymer matrices*. Revista Mexicana de Ingenieria Quimica, 7(3): 263-273.