



# UTILIZATION OF MANGO SEED STARCH IN MANUFACTURE OF HYBRID BIOCOMPOSITE REINFORCED WITH MICROPARTICLE ZnO AND CLAY USING GLYCEROL AS PLASTICIZER

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## ABSTRACT

Bioplastics are plastics that can be used just like conventional plastics, but will disintegrate by the activity of microorganisms into water and carbon dioxide. Starch is a natural polymer material that can be used for bioplastic production. The addition of reinforcing particles has been shown to improve the mechanical properties of bioplastics. The aim of this research is to know the potency of mango seed and microparticles of ZnO and clay as filler and glycerol concentration on tensile strength and elongation at break, functional group (FTIR) and surface morphology (SEM). In this study used mango seed starch size of 5 grams, with variation of ZnO mass of 0; 1; 3; and 6% wt, with variation of clay filler mass of 0; 3; 6 and 9 wt%, while mass of glycerol with a variation of 0; 20; 25; 30; and 35% wt. The FTIR analysis shows that no new functional groups are formed. From the analysis of mango starch content obtained 62.82%, 44.0% amilopectin content, amylose content 14.82%, and water content 12.65%. In this study obtained bioplastics with the best conditions on the use of 3% ZnO, 6% clay and 25% glycerol, with a tensile strength of 5,657MPa, percent elongation at breakup 43.431%.

**Keywords:** bioplastic, clay, mango, starch, ZnO.

## 1. INTRODUCTION

Plastic products are often used by people in everyday life as packaging materials because they are lightweight, relatively inexpensive, flexible, and practical. However, the use of plastic has disadvantages because it is a synthetic polymer with the main raw material derived from petroleum which is limited in number and cannot be renewed, so that plastic waste is very difficult to decompose by microorganisms. As a result, more and more people are using plastic, causing buildup that causes pollution and environmental damage. Various efforts to reduce the impact of plastic waste have been carried out. In addition to the plastic recycling process, environmentally friendly plastics have also been developed. Plastics made from synthetic chemicals are replaced with raw materials that are easily broken down by microorganisms naturally into environmentally friendly compounds, called biodegradable plastics [1].

Renewable resources that are known to be able to make biodegradable plastics are starch and cellulose. An alternative for a low cost and renewable substrate has been proposed by using agriculture waste [2]. In addition to performance and abundant sources, starch from agriculture waste is the solution for an alternative. Starch as biodegradable polymer becomes reasonable material for the production of bioplastics because of its low cost [3].

Much research has been done to make bioplastics using several natural polymers such as proteins, starches, and bacteria [4]. However, the use of natural materials used is less effective. Raw materials for bioplastics originate from natural constituents such as polysaccharides (eg starch, cellulose, chitin and lignin), proteins

(eggelatine, casein and wheat gluten) and lipids (eg plant oils and animals fats) [1].

Mango is a very common tropical fruit usually found in South Asia, especially in Eastern India, China, Burma, Andaman Islands and America The middle. Content of mango seeds in different varieties ranging from 9% to 23% by weight of fruit and the core content of seeds ranges from 45.7% to 72.8% [5]. Mango seeds are rich in carbohydrates, fats, proteins and minerals. This mango seed starch shows oval-to-shaped granules ellipse, similar to starch granules of beans. Various properties of starch mango seeds are comparable to starch from corn, wheat, rice and potatoes and can effectively utilized as a source of starch. In addition, mango seeds have a high enough starch content that potentially as an alternative substitute material in the manufacture of bioplastics. Amylose levels in mango seeds are expected to provide optimal mechanical properties and amylopectin levels provide optimum stickiness [6].

In this study the fillers used were ZnO and organic clay with the size of microparticles which were then combined with mango seed starch as the matrix for bioplastic formation was biodegradable and had better mechanical properties. The addition of plasticizers into biodegradable film blends is necessary to overcome the fragility of films caused by high intermolecular forces [7]. The nano-scale filler greatly influences the properties of the composite produced and showed improvements in physical and mechanical properties when compared with other conventional materials. Antimicrobial activity of nanoparticles related to several mechanisms [8].

ZnO (Zinc Oxide) nanoparticles have been widely known among researchers due to their use in



various applications such as gas sensors, chemical sensors, biosensors, superconductors, photo catalysts, optoelectronic devices, cosmetics, etc. Zinc Oxide is environmentally friendly and easy to be synthesized, have a stable nature, and are antibacterial [5]. Clay material is the material that attracts the most attention because it is strong, rigid, abundant in nature, cheap and capable of high interpreting particles into their structures. This intercalation is due to the small load of the layer so that the cations in the space between layers can be exchanged. In contrast to ordinary polymer composite materials, polymer-clay nanocomposites are formed if the polymer can be intercalated into the mineral clay gallery so the properties of the polymers are formed differ from the properties of the microparticles [8].

The addition of plasticizers plays a role in enhancing their plasticity, i.e. mechanical properties that are soft, resilient, and strong. Therefore, plasticization will affect physical properties and mechanisms films such as tensile strength, hardness elasticity, electrical properties, flow temperature, transition temperature glass, and so on [8].

Glycerol is obtained commercially as a by-product when fat and hydrolyzed oil to produce fatty acids or metal salts (soap). Glycerol is also synthesized on a commercial scale from propylene (obtained with cracking petroleum), because the supply of natural glycerol is inadequate. Besides synthesis by using propylene, glycerol can also be obtained during sugar fermentation sodium bisulfite when added to yeast [7].

Utilization of organic waste such as mango seeds for production of starch based bioplastic can help reducing the environmental damages that are caused by conventional plastics. Higher value bioplastics can be obtained by improving their properties with the most abundant and biodegradable reinforcing filler like cellulose. The goal of this work is to study the properties of biocomposite from agriculture waste. Effect of reinforcement fillers and plasticizers on biocomposite are also examined.

## 2. MATERIALS AND METHODS

Starch derived from mango seeds was obtained from juice traders in Medan, clay with particle size 10.813  $\mu\text{m}$ , and glycerol 99% was obtained from Rudang Jaya Medan.

### 2.1 Preparation Starch of Mango Seeds

Mango seeds (100 gram) obtained from waste container at local juice traders was washed with clean water before shredded to small pieces. The shredded mango seeds was later peeled and placed in mixing blender and soaked in water for about 100 ml. After mixing process, starch slurry was filtered and later placed in tank for settling that took at least 30 minutes. Starch sediment was separated from the slurry and then washed again with distilled water. After the second settling, starch sediment was dried using oven with temperature  $\pm 60^\circ\text{C}$  for removal of free water. Starch was sieved with strainer 100 mesh/inch for better homogeneous size [9] [10].

### 2.2 Biocomposite Preparation

Solution containing 0, 3, 6 and 9% wt/wt of fillers to starch was prepared by dispersing 100 ml distilled water and glycerol with concentration varied from 0, 20, 25, 30, and 35% wt/v of plasticizer to starch [11]. Solution was placed into ultrasonic homogenizer KUDOS tank and processed for about 50 minutes. After ultrasonication, solution was removed from the tank. Starch (5 gram) was added to the solution and heated using hot plate while stirred until it gelatinized ( $80.53^\circ\text{C}$ ). After mixing, the solution was cast onto flat and dried with temperature  $60^\circ\text{C}$  for 24 hour. Once set, bioplastic was cooled to ambient temperature before peeled off the flat.

### 2.3 Biocomposite Characterizations

Tensile strength was measured with GoTech Universal Testing Machine using the standard of ASTM D882-91. Tensile strength value was obtained from the observed data. Elongation at break is an indication of biocomposite flexibility and is expressed as a percentage.

### 2.4 Density

Density of biocomposite was investigated by the standard of ASTM D792-91 on film with size approximately 5x5 cm. The film's mass was measured using analytical balance. Density was calculated as follows:

$$\text{Density} = \frac{\text{mass(gram)}}{\text{volume}(\text{cm}^3)} \quad (1)$$

### 2.5 Water Uptake

Water uptake was investigated by cutting film with size approximately 2x2 cm and then weighed the mass. Film was put into a container filled with distilled water for 24 hour. After immersion in water, film was removed from the water and weighed to measure the wet weight. Water uptake was calculated as follows:

$$\text{Wateruptake} = \frac{\text{wetweight} - \text{dryweight}}{\text{wetweight}} \times 100 \quad (2)$$

### 2.6 Fourier Transform Infrared Spectroscopy (FT-IR)

Functional groups of biocomposite were analyzed by using IR Prestige-21 Shimadzu. The analysis using FT-IR represented spectrum data in graphic and wave numbers of each data that provided functional groups of bioplastics.

### 2.7. Scanning Electron Microscopy (SEM)

Surface morphology of biocomposite were analyzed by using Buck Scientific Model 500 Infrared Spectrophotometer. The analysis using SEM represented characteristics of fracture surface morphology from biocomposite.



## 2.8 Transmission Electron Microscopy (TEM)

The analysis using TEM represented particle size of zinc oxide (ZnO) as filler.

## 2.9 Gelatinization Profile Analysis Procedure with Rapid Visco Analyzer (RVA)

Gelatinization profile analysis of mango seed starch with RVA was carried out at Laboratory Services for Testing the Faculty of Agricultural Industry Technology, Padjadjaran University.

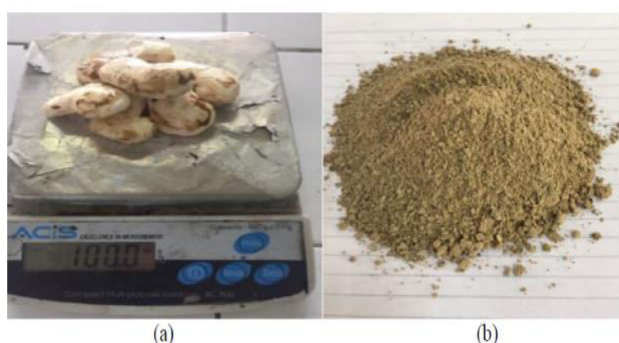
## 2.10 Biodegradability Test

The biodegradation test is carried out using soil burial techniques. Residual sample 10x10 cm placed and planted in a pot that has been filled with soil. Observation of the sample is conducted within a certain time frame.

# 3. RESULTS AND DISCUSSIONS

## 3.1 The Result Starch Extraction from Mango Seed

In this study, the raw materials for hybrid biocomposite is starch extracted from mango seeds. Mango seeds obtained from juice traders located at the Pembangunan and Padang Bulan, Medan. The resulting starch greyish powder form with a particle size  $\pm 100$  mesh. The results starch extraction from mango seeds yield of starch obtained by 43.2 %, which of 100 g dried starch mango seeds gained as much as 43.2 g and then further analysis of starch obtained.



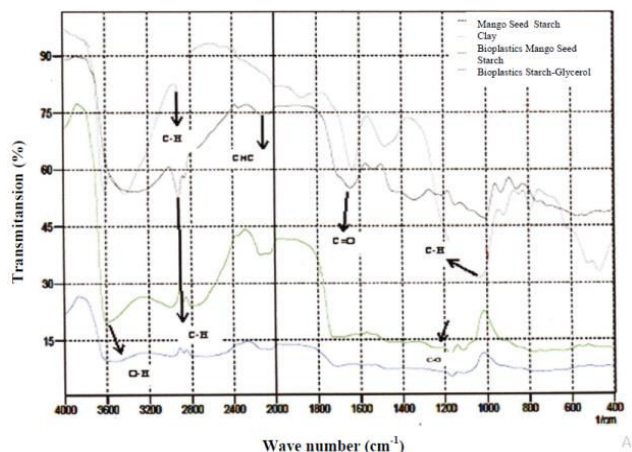
**Figure-1.** (a) The results extraction (b) starch the mango seeds of starch from mango seeds.

In this study, the composition of mango seed starch obtained can be seen in Table-1.

**Table-1.** Mango seed starch composition.

Parameter	Composition (%)
Amilopectin content	44
Amilose content	14.82
Water content	12.65
Starch content	75.47

## 3.2 Fourier Transform Infrared Spectroscopy (FT-IR)

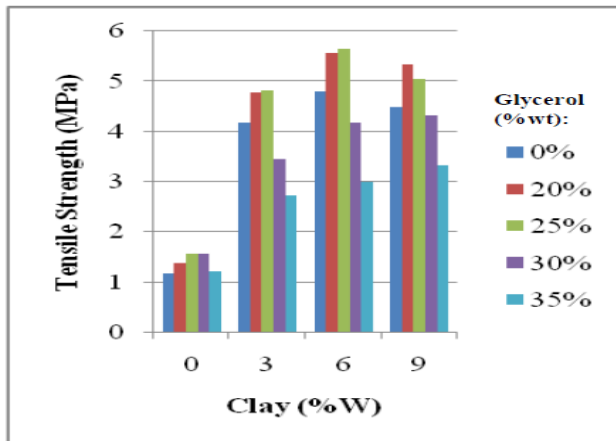


**Figure-2.** FTIR Specs of Mango Seeds Starch, Clay, Pure Starch Biocomposite, Strach/Clay/Glycerol Biocomposite and Starch/Glycerol Biocomposite.

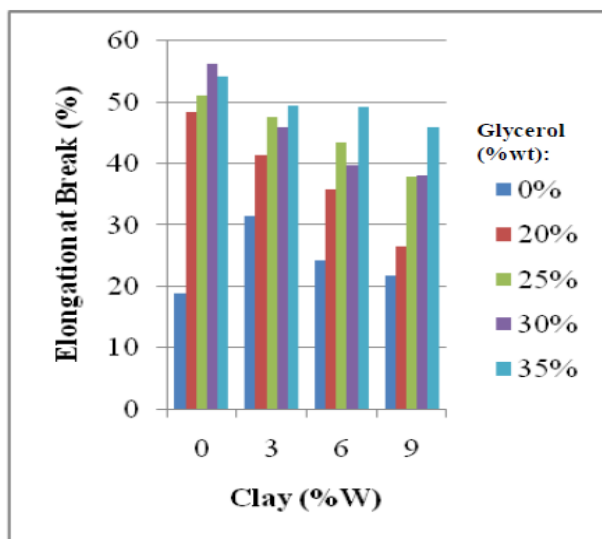
Based on Figure-2, the biocomposite starch of mango seed with glycerol plasticizer generated several peaks of wave numbers in each range of region. There is a peak of the wave number  $3527.02 \text{ cm}^{-1}$  to  $2677.20 \text{ cm}^{-1}$ . The peak corresponds to the absorption caused by the C-H bond (type alkane compound), and OH (type of phenol compound, hydrogen bonding alcohol). At peak with wave numbers  $2140.99 \text{ cm}^{-1}$  and  $2059.98 \text{ cm}^{-1}$ , corresponds to the absorption caused by the triple bonds of  $\text{C}\equiv\text{C}$  (alkaline type compounds). At the peak with the wave numbers  $1948.10 \text{ cm}^{-1}$  and  $1550.77 \text{ cm}^{-1}$ , corresponding to the absorption caused by the bonding  $\text{C}=\text{O}$  (type of aldehyde, ketone, carboxylic acid, esters) and  $\text{C}=\text{C}$  (type of alkene compound). At peak with wave numbers of  $1477.47 \text{ cm}^{-1}$  to  $459.06 \text{ cm}^{-1}$ , corresponds to absorption caused by the C-H bond (alkane type compound), and C-O (type of alcohol, carboxylic acid, ester). Bioplastic mango starch starch has a functional group which is a combination of specific functional groups contained in its constituent components such as C-H, O-H, C-O,  $\text{C}\equiv\text{C}$ ,  $\text{C}=\text{O}$ , and  $\text{C}=\text{C}$ . The wavelength of the functional group OH in clay differs from the bioplastic starch of mango seed with the addition of glycerol plasticizer wherein the addition of glycerol causes the functional group OH to shift with wave length  $3622.32 \text{ cm}^{-1}$  to  $3522.02 \text{ cm}^{-1}$ . It identifies that there is a process of forming new bonds between matriks mango starch and glycerol so as to weaken the OH bond on the matrix of mango seed starch. In bioplastics with the addition of glycerol, the C-O and OH groups form a random carbon chain, thus causing the sample to become more elastic. Chemical interactions are reflected by changes in the characteristics of the absorption peak spectrum after mixing of two or more matrix intermediates with added chemicals such as clay and glycerol [6].



### 3.3 Tensile Strength and Elongation At Break



**Figure-3.** Tensile strength of starch based biocomposite reinforced with clay and glycerol as plasticizer.



**Figure-4.** Elongation at break of starch based biocomposite reinforced clay using clay as plasticizer.

In Figure-3 and Figure-4 can be seen the effect of adding clay fillers and glycerol plasticizers to tensile strength and elongation at breaks from biocomposite. The highest bioplastic tensile strength value was obtained in addition of 3% clay filler and 30% glycerol plasticizer with 5.657 MPa value. For the elongation value at the highest breakup bioplastic was obtained on the addition of 6% clay filler with value of 47.553%. While in addition of glycerol elongation value at breakup biocomposite at concentration 35% glycerol with value 49.139%.

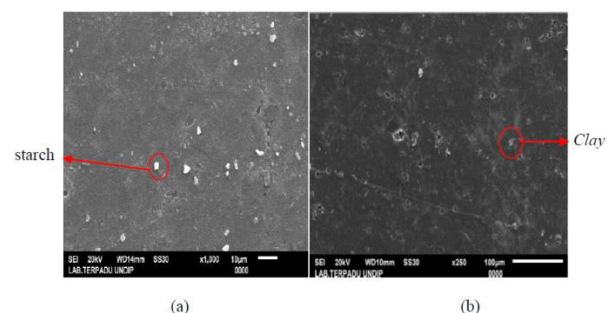
The influence of the addition of organic clay causes more hydrogen bonds in the biocomposite so that the chemical bonds are stronger and harder to break because they require a large amount of energy to break the bond. While the percentage elongation is inversely proportional to the addition of natural clay. Where the natural addition of clay, causing percentage elongation decreased. This is due to the decreasing intermolecular

bonding distance. Clay has a micron-size surface area large enough to be able to interact effectively with the polymer matrix at low concentrations (5-8%). As a result, polymer clay showed an increase in modulus, thermal stability, and barrier properties without an increase in density and loss of optical properties.

In addition the high mass of clay filler can contribute to a slowdown in the interaction between molecules of starch bioplastics. This leads to the development of bioplastic structures becoming heterogeneous and the results being discontinuities. With the addition of glycerol as plasticizer, plasticizer molecules in bioplastics located between biopolymer bonding chain and be able to interact by forming hydrogen bonds in the bond between the polymer chains, causing interactions between the molecules of biopolymers have become less and may cause a decrease in biocomposite material stiffness. This is due to the increased speed of viscoelastic response and molecular mobility of the polymer chain. The addition of plasticizers serves as a conduit on the elastic properties of bioplastics, so more plasticizer provided will increase the value of the plastic extension. Plasticizer can reduce internal hydrogen bonds of molecules and lead to weakening of the intermolecular attractive force adjacent polymer chains, thereby reducing the tensile breaking. Plasticizer can reduce internal hydrogen bonds of molecules and lead to weakening of the intermolecular attractive force adjacent polymer chains, thereby reducing the tensile breaking. Based on this, it can be seen that elongation percentage is inversely proportional to tensile strength [5].

### 3.4 Analysis Morphological Characteristics Fault Biocomposite Starch Extraction Mango Seeds with Clay as Filler and Plasticizers Glycerol

Characteristics of fracture surface morphology is shown by analysis Scanning Electron Microscopy (SEM). Presented below are the characteristics of the results fracture morphology analysis bioplastics with the composition of the starch content of skin 5 grams of mango seeds, clay 6 % and 25 % gliserol at 1000x magnification. Characteristics SEM morphology analysis carried out in the laboratory of SEM, University of Diponegoro.



**Figure-5.** Morphology analysis report fault (a) Biocomposite mango seeds starch and (b) Biocomposite from mango seeds starch with microparticles clay as filler and glycerol plasticizer in magnification 1000x.



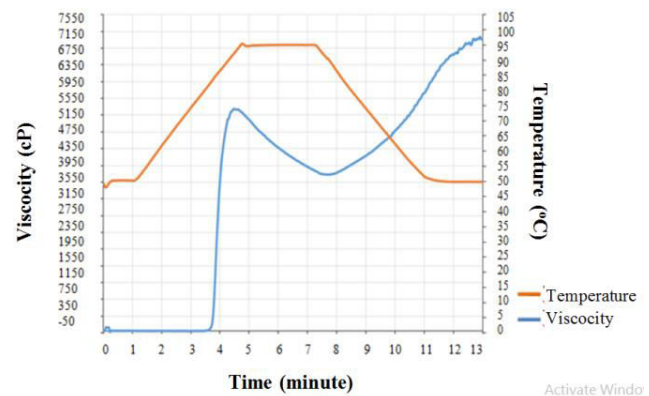


From Figure-5 it can be seen analysis fracture morphology, strength testing. The attraction of bioplastics. On the results of SEM analysis of mango seeds starch bioplastics without filler microparticle clay and glycerol (Figure-5 (a)) shows are clumps of insoluble mango seed starch during plastic film making. It identifies that the starch in the matrix is poorly dissolved but is spread fairly evenly. The lack of sufficiently strong power in the heating and stirring process between the filler and the starch matrix is likely to cause the starch blob not fully dissolved properly. If there is a sufficiently strong force such as good agitation in the blending process at gelatinization temperature and glass transition will easily incorporate insoluble starch particles resulting in better distribution [5].

While at biocomposite mango seeds starch with microparticle clay and glycerol (Figure-5(b)) SEM analysis results show white spots that are clay microparticle fillers and describe the clay particle size distribution. The above results show a fairly uniform dispersion of clay fillers into the matrix. The uniform dispersion shown from the SEM results shows that there is good adhesion between the micro-sized filler and the matrix. In accordance with research Azizi *et al.* (2014) which states that clay films in matrices are uniformly dispersed and the uniform distribution of micro-filler in matrices plays an important role in improving mechanical properties. The ultrasonic process of clay microparticles is intended to produce materials with unusual properties, which lead to the formation of larger particle surfaces, smaller particle size [5]. On the surface of the bioplastic morphology does not show any empty fraction. This is due to the influence of microparticle-sized clay fillers so that the structure is more compact [6]. From the above SEM analysis it can be concluded that the addition of clay microparticles dispersed well enough in the starch matrix is indicated by the optimum result of bioplastic mechanical characteristics with tensile strength value 5.657 MPa, percent extension at break off 43.431%, density 1.315 gr/cm<sup>3</sup> and percent Water absorption 32.282%

### 3.5 Characteristics Gelatinization of Starch with RVA (Rapid Visco Analyzer)

The characteristics of the gelatinization profile of mango seed starch were measured by RVA (Rapid Visco Analyzer). This characteristic is related to the measurement of starch viscosity certain concentrations during heating and stirring.

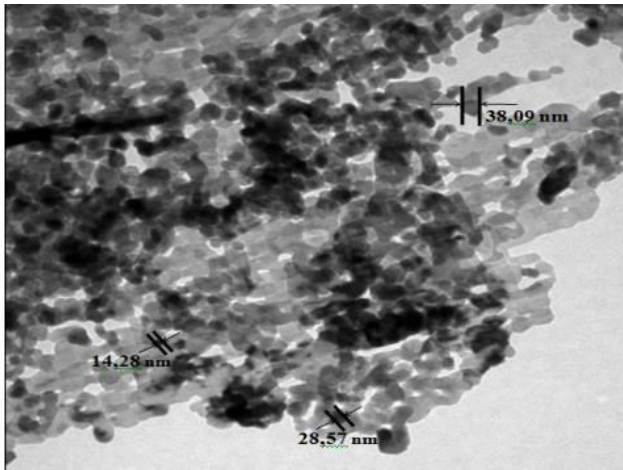


**Figure-6.** Graph of relationship between time and temperature on starch viscosity mango seeds.

Based on figure-6, it can be seen that the peak viscosity achieved is 5303 cP. On heating above 80.53 °C, mango seed starch experiences the decrease in viscosity is quite sharp with a breakdown viscosity of 1577 cP. Mango seed experiences an increase in viscosity during the cooling phase. The viscosity of the setback during this cooling phase is 3240 cP. Setback viscosity mango seed starch is relatively high, which shows the tendency of mango seed starch more easily experience retrogradation. Like starch in general, mango seed starch has a gelatinization profile with a high peak viscosity and is followed with a sharp decrease in viscosity during the heating phase. This is because the warming that causes the kinetic energy of water molecules becomes stronger of the attractive attraction between starch molecules in the granules, so that water can enter the starch and starch will expand [12].

### 3.6 Characterization of Transmission Electron Microscope (TEM) ZnO (Zinc Oxide) Nanoparticle Filler

Purpose of the characterization of the Transmission Electron Microscope (TEM) filler ZnO nanoparticles (Zinc Oxide) is to determine the shape and size ZnO nanoparticles (Zinc Oxide) obtained from Alibaba.com. TEM characteristics ZnO nanoparticles are shown in Figure-7.



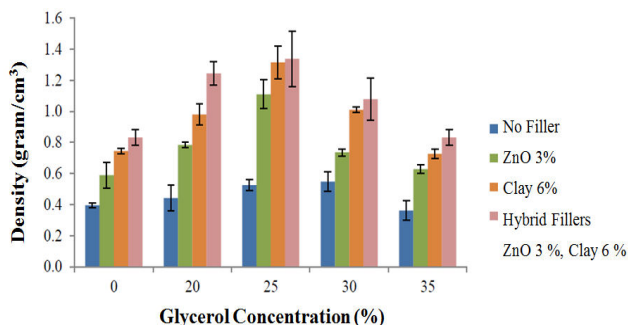
**Figure-7.** Characteristics of Transmission Electron Microscope (TEM) Fillers ZnO nanoparticles (Zinc Oxide) at 200 nm magnification.

Figure-7 above is the shape and size of the ZnO nanoparticles produced spherical (Spherical-Shaped) with various diameter sizes particles namely 14.28; 28.57 and 38.09 nanometers so the average diameter size nanoparticles smaller than 30 nanometers.

Nano size is usually measured in nanometers (1 nm is equivalent to 10 m) and that includes systems whose size is above the molecular dimensions and below macroscopic dimensions (usually > 1 nm and <100 nm) [13]. So fillers ZnO nanoparticles (Zinc Oxide) can be classified as sized fillers nanometer.

### 3.7 Effect of Addition to Hybrid Fillers with Glycerol Plasticizer against Biocomposite Density

Figure-8 shows the effect of adding fillers ZnO and clay hybrids with glycerol plasticizers on biocomposite densities from mango seed starch.



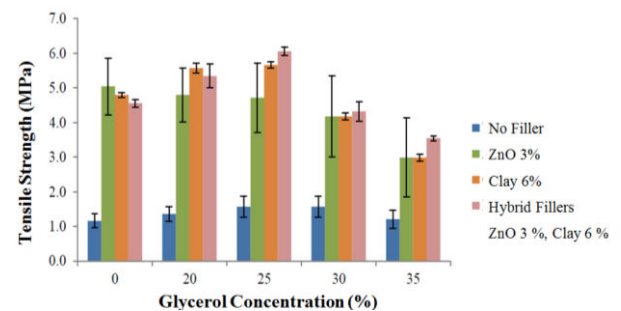
**Figure-8.** Effect of addition of ZnO, Clay, hybrid and glycerol plasticizer fillers against biocomposite density.

In Figure-8 can be seen the effect of adding hybrid and filler glycerol plasticizer against the biocomposite density of mango seed starch. In the chart above it can be seen that the addition of ZnO and clay as fillers and glycerol as plasticizers produce density values tend to increase. Density Value the highest was obtained in biocomposites with the addition of 3% ZnO and hybrid fillers 6% clay and 25% glycerol concentration with a

density value of 1.338 gram / cm<sup>3</sup>. While the lowest density value is obtained in biocomposites with no addition of 35% filler and glycerol by 0.362 gram/cm<sup>3</sup>. Density is proportional straight with the mass of a material, so the greater the mass of a material then the greater the density value. Addition of zinc oxide and organic inorganic fillers clay can increase the density of plastic mass, but at a certain point resulting in molecules between the components of the biocomposite are not dense [14].

### 3.8 Effect of Addition of Hybrid and Plasticizer Glycerol Fillers on Properties of Biocomposite Tensile Strength

Figure-9 shows the effect of adding hybrid fillers ZnO and clay with glycerol plasticizers on tensile strength properties biocomposite from mango seed starch.



**Figure-9.** Effect of addition of ZnO, clay, hybrid and plasticizer fillers glycerol on the properties of biocomposite tensile strength.

In Figure-9 we can see the effect of adding hybrid fillers and glycerol plasticizer against the tensile strength of biocomposites made with using mango seed starch. In the picture above it appears that the addition glycerol concentration to tensile strength tends to increase then decrease when adding glycerol concentration by 30%. Highest tensile strength value obtained in biocomposites with the addition of 3% ZnO hybrid filler and 6% clay and 25% glycerol concentration with a value of 6.052 MPa. While the value of tensile strength the lowest is obtained in biocomposites without the addition of fillers and without glycerol with a value of 1.158 MPa. The value of the tensile strength in the biocomposite that uses hybrid fillers are lower when compared to the addition of ZnO or clay only, but when the glycerol concentration is added by 25%, the tensile strength of hybrid fill biocomposites is higher than other biocomposites.

From the results of this test, it shows that the addition of ZnO and clay fillers glycerol concentrations in biocomposites have different effects on the tensile strength of the biocomposite produced [13]. The addition of ZnO and clay fillers, the tensile strength value the higher the biocomposite produced. This is due to increasing many ZnO and clay fillers will affect the structure of the biocomposite [15].

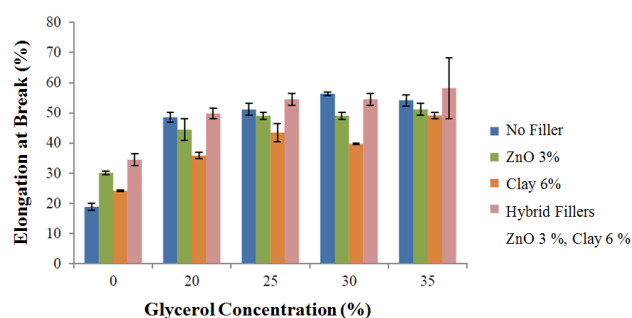
The results above show that more glycerol is added the nature of the tensile strength will be lower, but



if glycerol is added too little, the biocomposite produced will be easy to experience fracture / less elastic [16]. With the addition of glycerol as a plasticizer, plasticizers in the biocomposite are located between bonding chains biopolymers and can interact by forming hydrogen bonds in chains bonding between polymers thereby causing interactions between biopolymer molecules becomes less and less. Addition of glycerol can reduce strength intermolecular biocomposites between polymer chains and increase flexibility biocomposite [14].

### 3.9 Effect of Addition of Hybrid and Plasticizer Glycerol Fillers on Elongation when Disconnecting Biocomposites

Figure-10 which shows the effect of adding fillers glycerol hybrids and plasticizers against elongation when breaking up biocomposites.

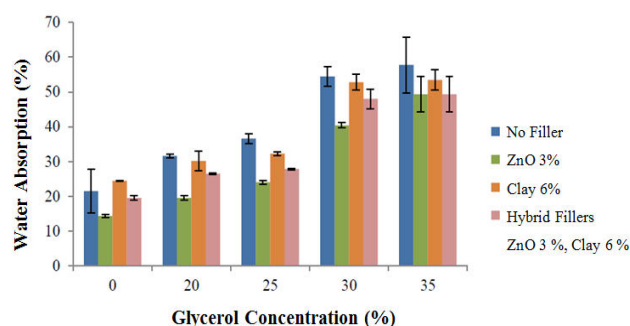


**Figure-10.** Effect of addition of ZnO, clay, hybrid and plasticizer fillers glycerol against elongation when biocomposite disconnect.

In Figure-10 we can see the effect of adding hybrid fillers and glycerol plasticizer against elongation at the time of breaking up of the biocomposite that is made by using mango seed starch. In the picture above it appears that the addition of glycerol concentration to elongation at the time of breaking up tends increased. The elongation value at the time of highest breaking is obtained at biocomposite with the addition of 3% ZnO and 6% clay fillers as well glycerol concentration 35% with a value of 58.148%. While the elongation value at when the lowest break is obtained in biocomposites without the addition of fillers and without glycerol with a value of 18.775%. The value of the tensile strength in the biocomposite using a hybrid charger is higher when compared to additions only ZnO or clay fillers. From the results of this test, it shows that the addition ZnO fillers and clay glycerol concentrations in biocomposites have an effect different at elongation at the time of breaking up from the resulting biocomposite [14]. The elongation value at the time of break shows an upside down tendency with a tensile strength test. Where the addition of fillers, causes value elongation at the time of breaking up decreases. This is caused by increasingly decreasing intermolecular bonding distance [17].

### 3.10 Effect of Addition of Hybrid Fillers and Glycerol Plasticizer on Properties of Biocomposite Water Absorption

Figure-11 which shows the effect of adding fillers glycerol hybrids and plasticizers on the absorption properties of biocomposite water.



**Figure-11.** Effect of addition of ZnO, clay, hybrid and plasticizer fillers glycerol on biocomposite water absorption properties.

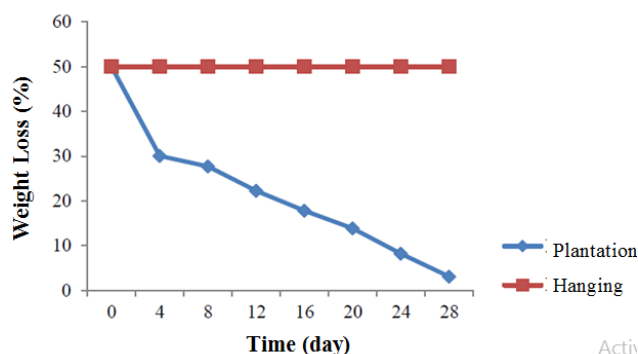
In Figure-11 shows the test results of water absorption for biocomposites by using hybrid fillers, as well as glycerol plasticizers. The lower percent water absorbability is obtained, the better the quality of the plastic. On the results above show that the addition of glycerol plasticizer tends to increase water absorption value. The highest absorption value of water is obtained in biocomposites without the addition of fillers with 35% glycerol concentration of 57.737%.

While the lowest biocomposite water absorption value is obtained on addition ZnO filler 3% and without the addition of glycerol with a value of 12.990%. Water absorption is the amount of water absorbed by the plastic film in percent after the test sample soaked in water at room temperature for 24 hours. The water fills the spaces blank in the plastic film. From the results obtained it can be seen that addition of glycerol plasticizer increases the value of water absorption. This increase due to the fact that glycerol is a hydrophilic compound and is also a nature compatible with starch matrices [16].

Water absorption starch biocomposites with ZnO fillers decreased when the addition of ZnO fillers. This is due to the value of water absorption with a smaller ZnO metal amplifier compared to the natural reinforcement of clay and starch itself because of the metal reinforcement has better resistance than natural boosters composed of polysaccharides [17].

### 3.11 Biodegradability Test of Mango Seeds with Hybrid Fillers and Gliserol Plasticizers

Presented below are the results of the biocomposite biodegradability test from seed starch mangoes with hybrid fillers and glycerol plasticizers.



**Figure-12.** Biodegradability test of mango seed starch biocomposites with fillers glycerol hybrid and plasticizer.

Figure-12 is the result of biodegradability test of mango seed starch biocomposite with a hybrid filler and glycerol plasticizer while biocomposite biodegradability test results using 3% ZnO concentration and clay 6%. The test is carried out on 3 samples for each variation. Reduction of sample mass is seen every four days for 28 days. This method done by planting the sample in the soil and hanging the sample on the air then calculates the residual weight of the sample in each unit of time (gram/day).

It can be seen that the reduction in residual weight for the treatment by planting is faster than the treatment by planting hanged up. This is because in the soil there are microbial activities that occur on biocomposites, causing degradation more quickly if compared to treatment by hanging [13]. Bacteria that are in the soil will decompose the plastic that has been planted so that it breaks polymer chains become monomers through the resulting enzyme from these bacteria. The ability of biocomposites to degrade due to chain shortness the lower the bond the more molecular weight the easier the polymer degraded [18].

#### 4. CONCLUSIONS

The mango seeds starch/ZnO/Clay bioplastics increased in tensile strength from 1.567 MPa for pure starch bioplastic to 5.797 MPa. Contrary to tensile strength, a decrease in elongation at break was reported. The improvement in bioplastics with reinforcing ZnO and clay could be attributed to the strong hydrogen bond between hydroxyl groups of the interface of both ZnO and clay fillers and starch matrix. This formation is also influenced by the percolation mechanism. Density of bioplastics increased as ZnO and clay content increased. Furthermore, addition of ZnO and clay content decreased water uptake of bioplastics. The incorporation of ZnO and clay particles into starch matrix resulted in agglomerates that caused deflections in bioplastics. FT-IR spectra provided information about hydrogen bond through its characteristic peak. Bioplastic with the highest tensile strength assumed to have better characteristics than the other bioplastics was found at ZnO content 3%, clay content 6% and 25% glycerol.

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