

Utilization of Sugar Palm (*Arenga pinnata*) Dreg in Biodegradable Plastic Processing

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ABSTRACT: Bioplastics are made from biomass sources that can be decomposed naturally in a relatively short time compared to plastics produced from synthetic polymers plastic. Sugar palm dregs are a promising source for bioplastics due to their abundance and renewability, and they do not compete with human needs. Sugar palm dregs contain enough crude fiber (41.66%) to produce strong bioplastics. Chitosan and glycerol are added to improve the performance of bioplastics. The responses observed included tensile strength, elongation at break, young modulus, thickness, biodegradability, and water vapor permeability. This study aims to determine the factors' effect on the response parameters and find the optimal multiresponse combination to fabricate sugar palm dreg-based bioplastics. The experimental design was determined using Taguchi method, and multiresponse analysis was carried out using the Grey Relational Analysis approach. The results show that adding sugar palm dreg increased the tensile strength, Young modulus, and thickness of bioplastics but decreased the elongation at break. Adding chitosan affected the water vapor permeability, and glycerol increased the biodegradation percentage of bioplastics. The optimal combination of sugar palm dreg bioplastic was 3 grams of sugar palm dreg, 2 grams of chitosan, and 3 grams of glycerol. The combination resulted in a tensile strength of 1.46 MPa, 24.49 of elongation at break, 6.08 MPa of young modulus, 0.28 mm of thickness, 100% of biodegradation, and 0.61 g.mm/kPa.hour.m². The results show that sugar palm dreg bioplastic is potential as future food packaging.

Keywords: bioplastic; sugar palm dregs; Taguchi method, Grey Relational Analysis

INTRODUCTION

Plastic waste becomes an international issue because it is difficult to decompose naturally then can cause environmental pollution both on land and water. Jambeck *et al.* (2015) reported that in 2010, 275 million metric tons of plastic waste was generated in 192 coastal countries, with 4.8 to 12.7 million metric tons entering the ocean. It causes environmental pollution if not balanced with proper waste management. The environmental issues have led to the creation of new breakthroughs in the packaging field, which is called biodegradable plastics (bioplastics). Bioplastics are plastics made from biomass sources so they can decompose naturally in a relatively short time compared to synthetic polymers. Bioplastics are made from biomass sources which are usually found in plants, such as starch, lignin, and cellulose, or in animals, like casein, chitin, chitosan, and so on (Harsojuwono & Arnata, 2015).

The research on bioplastics has been developed in the previous period. Bioplastics fabrication utilizes various types of materials, such as rice with chitosan and glycerol (Martina *et al.*, 2016), rice straw (Pratiwi *et al.*, 2016), cassava dreg (Nurlita *et al.*, 2017), corncobs (Wiradipta *et al.*, 2017), cassava starch reinforced with palm fiber and cassava fiber (Edhirej *et al.*, 2017), betel nut husk (Tamiogy *et al.*, 2018), and corn starch reinforced with

sugar palm fiber and corn husk (Ibrahim *et al.*, 2020) that added by chitosan and glycerin to improve their strength. Maltodextrin is the one of polysaccharides that is mostly used for coating material. Maltodextrin is a coating that can increase the encapsulation efficiency, this is probably because of the ability of maltodextrin to form the encapsulant surface during the drying process (Balasubramani *et al.*, 2015).

Sugar palm dreg is another agricultural material that can be used in bioplastics fabrication. Sugar palm dreg is a waste from the processing of sugar palm starch. Klaten, Jawa Tengah is one region that produces large quantities of sugar palm dreg. According to Styana and Hindarti (2017), there were 137 sugar palm starch producers with an average annual production of 200 tons. From those sugar palm starch productions, solid waste in the form of fibers will be produced for 659 tons per year. Sugar palm dreg contains biomass sources such as lignin, cellulose, and hemicellulose. Based on Purnavita and Sriyana (2013), sugar palm dreg contains 14.21% lignin, 60.61% cellulose, and 15.74% hemicellulose. Ilyas *et al.* (2017) state that sugar palm fiber (SPF) is a natural lignocellulosic fiber characterized by high resistance to seawater, high tensile strength, low degradation rate, and durability.

Therefore, this study will utilize sugar palm dreg in the fabrication of bioplastic mixed with chitosan and glycerol. The use of sugar palm dreg in this study was expected to improve the mechanical properties of bioplastics, while chitosan used as a stabilizer, thickener, binder for materials in making glass, plastic, and rubber, improve the mechanical properties and resistance of bioplastics to water (Rosida *et al.*, 2018). Glycerol used as a plasticizer to improve the elasticity of bioplastics. This study aims to determine the factors' effect on the response parameters and find the optimal multiresponse combination in the fabrication of sugar palm dreg-based bioplastics.

Microencapsulation can be prepared by the spray drying method. Technically, spray drying is a technique of changing samples from liquid to powder particles by spraying feed into a hot drying medium. In recent years, spray drying techniques have been developed for the encapsulation process of food compound components, such as bioactive content, flavor, and also antibacterial compounds. Microencapsulation using glucomannan as the coating material has been reported to be able to protect bioactive compounds in spray drying encapsulation (Wattanaprasert *et al.*, 2016). This study aimed to study the encapsulation efficiency of microencapsulation of sweet potato leaf extract through the combined amount of maltodextrin and glucomannan in a microencapsulation agent using the spray drying method.

MATERIALS AND METHOD

Sugar Palm Dreg Sample

The materials used in this research are sugar palm dreg obtained from a local farm in Klaten, Jawa Tengah, Indonesia. Glycerin (ACS reagent, ≥ 99.5%), chitosan (94.88%), acetic acid, and aquadest were purchased from CV. Chemix Pratama (Indonesia).

Design Experiment

The design experiment of this research was determined by an orthogonal array of Taguchi method. The factors in this research consist of sugar palm dreg (A), chitosan (B), and glycerol (C). Each factor has three levels factor. The

orthogonal array used for the design experiment is L₉(3⁴) as shown in Table 1.

Sugar Palm Dreg Preparation

Sugar palm dreg in fiber form were cut into pieces approximately 2 cm in length. The fibers were dried by oven dryer at ±105 °C for 6 hours, grinded, and then sieved through a 200 mesh sieve.

Production of Bioplastics

Bioplastics are made by solution casting techniques. Sugar palm dreg and glycerol as plasticizers were blended in 50 ml of distilled water using a magnetic heated stirrer (S/N 05150, HMS-79, China) at 60 °C for 10 minutes. The chitosan was dissolved in 100 ml of 1% acetic acid at 60 °C for 30 minutes and then added to the sugar palm dreg and glycerol solution. The mixture was mixed by using a magnetic heated stirrer for 15 minutes. Then, the bioplastics solution was poured into the mold and placed at room temperature for 3–4 days.

Analytical Method

Mechanical Characterization (Tensile Strength, Elongation at Break, Young Modulus)

Mechanical properties of bioplastic were measured according to ASTM D882 (2002) standard using Universal Testing Machine (S/N 153409/2002, Model Z05, ZwickRoell, GmbH & Co.KG, Jerman). The testing started by mounting the sample specimen firmly between the tensile clamps. Then, the clamps would pull the samples till they broke off. The result of the measurement will appear in the censor instrument that was connected to the tensile machine. The measurements were conducted for two replications of each specimen.

Physical Characterization (Thickness and Biodegradation Percentage)

Bioplastic thickness was measured using a micrometer screw gauge accuracy of 0.01 mm (Trickle 0-25 x 0.01 mm, China). The measurements were conducted on five different random positions for each sample. Meanwhile, the biodegradation percentage was determined by the soil

Table 1. The design experiment of L₉(3⁴) orthogonal array

Experiment	Level factor ratio			The amount of each factor		
	A	B	C	Sugar palm dreg (g)	Chitosan (g)	Glycerol (g)
1	1	1	1	1	1	1
2	1	2	2	1	1.5	2
3	1	3	3	1	2	3
4	2	1	2	2	1	2
5	2	2	3	2	1.5	3
6	2	3	1	2	2	1
7	3	1	3	3	1	3
8	3	2	1	3	1.5	1
9	3	3	2	3	2	2

Note: (A) factors of sugar palm dreg, (B) factor of chitosan, (C) factor of glycerol

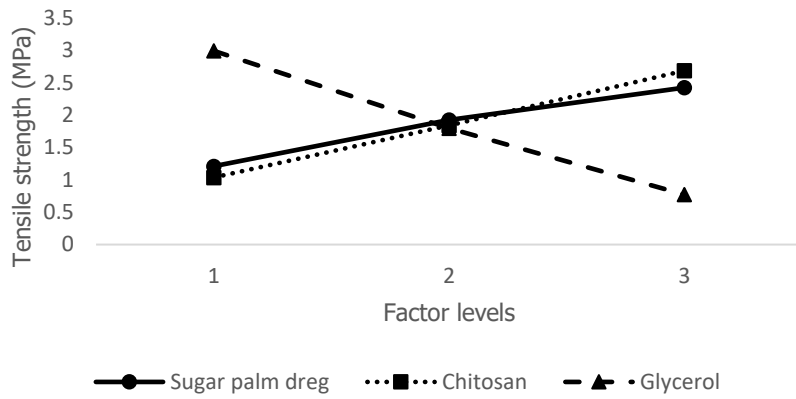


Figure 1. The effect of the factors on tensile strength

burial test. Samples 2.5 cm x 4 cm were conditioned in the desiccator until they reached constant mass and were noted as the initial mass. Then, the samples were placed in the plastic glass and buried with soil at 5–10 cm depth for 12 days.

Barrier Characterization (Water Vapor Permeability)

The test was performed according to ASTM E96 (1989) standard with a slight alteration according to Siah *et al.* (2015). Silica gels were heated at 105 °C for 3 hours. Samples were sealed properly on a circular cup filled with 15-gram heated silica gels. The cups were placed in a desiccator with distilled water and weighed at 1–6 hours intervals.

RESULT AND DISCUSSION

Before being used in bioplastic fabrication, sugar palm dreg was first subjected to a proximate test to determine its chemical composition. The proximate test results show that the sugar palm dreg has a moisture content of 0.93%, 0.06% fat, 41.66% crude fiber, and no protein.

Mechanical Properties

This study's mechanical characterization of bioplastics included tensile strength, elongation at break, and young modulus. The results of the measurement of the mechanical response parameter are shown in Table 2.

Tensile strength is the maximum load or stress which bioplastics can withstand. The greater the tensile strength, the greater ability of the bioplastic to withstand mechanical loads. The results in Figure 1 demonstrate that increasing the concentration of sugar palm dregs and chitosan leads to higher measured tensile strength. It was because sugar palm dreg contained crude fiber (41.66%), which made the bioplastic strong and stiff. Naturally, cellulose molecules are arranged in the form of fibrils which consist of several cellulose molecules linked by hydrogen bonds. These fibrils form a crystal structure covered with lignin. Such chemical composition and structure made most materials containing cellulose strong and hard (Sari *et al.*, 2018). Meanwhile, the higher concentration of glycerol, the smaller the tensile strength value measured. It is because glycerol, as a plasticizer, can make bioplastics elastic. According to Hardjono *et al.* (2016), plasticizers can reduce intermolecular forces, widen the distance between molecules, and increase plastic elasticity. The increase in elasticity will impact the decreased tensile strength value.

Elongation at break is the elongation of the bioplastic measured right before it breaks off. The higher elongation at the break value, the more ductile the bioplastics. The results in Figure 2 show that elongation at break is inversely proportional to tensile strength. The higher the concentration of sugar palm dreg, the smaller the

Table 2. Mechanical properties of the bioplastic

Experiment	Tensile Strength (MPa)	Elongation at break (%)	Young Modulus (MPa)
A1B1C1	1.53±0.39	6.10±2.77	30.9±22.00
A1B2C2	1.11±0.164	10.65±0.12	10.36±1.43
A1B3C3	0.99±0.19	11.76±0.28	8.39±1.39
A2B1C2	1.08±0.32	4.74±0.886	24.37±12.33
A2B2C3	0.83±0.23	6.70±1.25	12.32±2.06
A2B3C1	3.87±0.85	3.90±0.62	100.71±25.99
A3B1C3	0.49±0.28	4.59±1.32	11.04±6.18
A3B2C1	3.58±0.19	2.71±0.71	139.82±45.55
A3B3C2	3.20±0.46	5.62±2.04	66.15±38.55

Note: (A) factors of sugar palm dreg, (B) factor of chitosan, (C) factor of glycerol. The numbers following the letters are the level factor ratio.

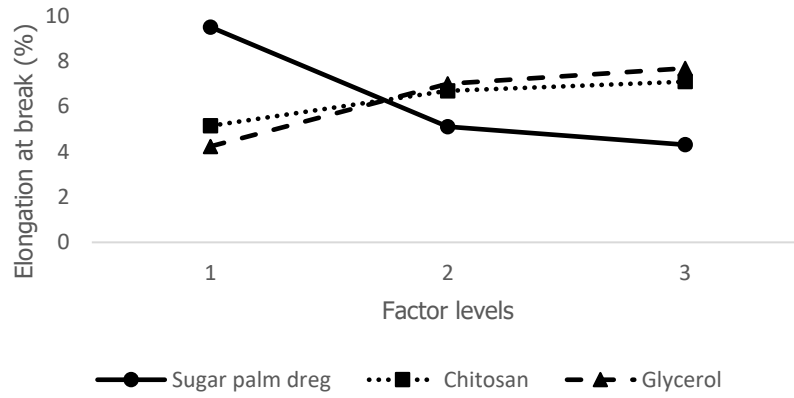


Figure 3. The effect of the factors on young modulus

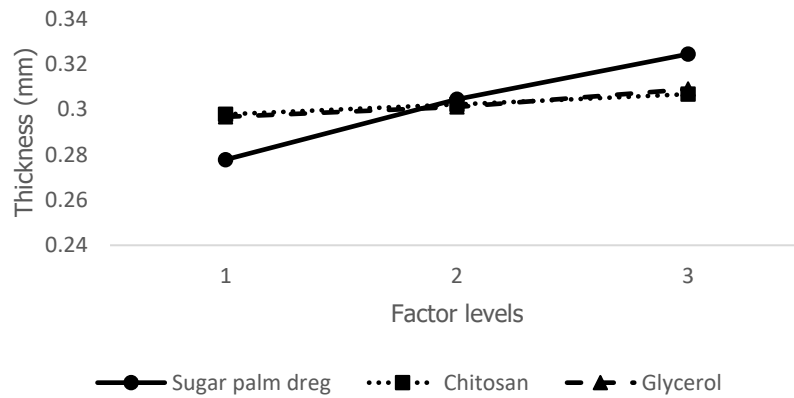


Figure 4. The effect of the factors on the bioplastics thickness

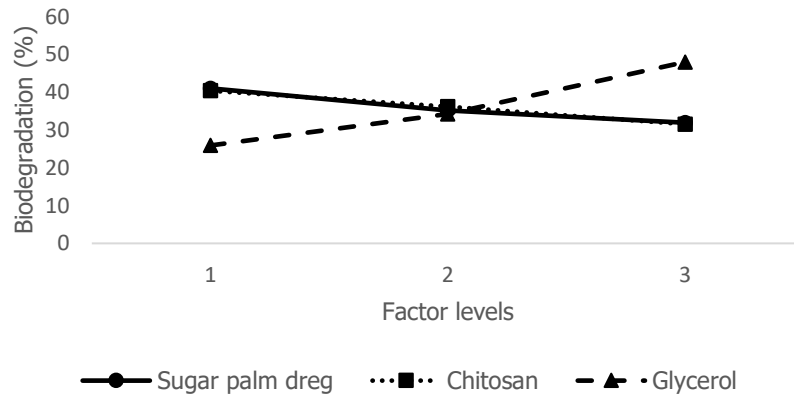


Figure 5. The effect of factors on biodegradation percentage

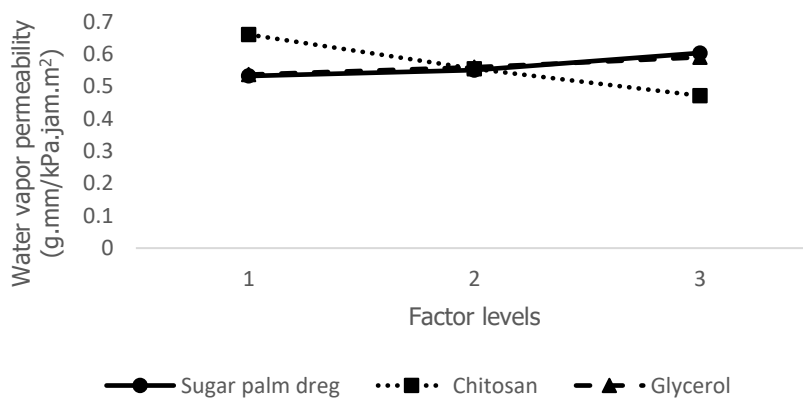


Figure 6. Effect of the factors on water vapor permeability

Table 3. Physical properties of the bioplastic

Experiment	Thickness (mm)	Biodegradation (%)
A1B1C1	0.27±0.01	33.61±12.46
A1B2C2	0.28±0.01	41.58±10.40
A1B3C3	0.29±0.00	47.80±6.84
A2B1C2	0.30±0.00	37.05±12.69
A2B2C3	0.31±0.00	45.61±8.11
A2B3C1	0.30±0.01	22.75±5.99
A3B1C3	0.33±0.00	50.45±14.10
A3B2C1	0.32±0.00	21.33±4.36
A3B3C2	0.33±0.01	23.99±6.49

Note: (A) factors of sugar palm dreg, (B) factor of chitosan, (C) factor of glycerol. The numbers following the letters are the level factor ratio.

elongation value obtained and the greater the bioplastic elongation value, along with adding chitosan and glycerol.

Young modulus can be interpreted as a measure of the stiffness of the plastic. The greater the young modulus, the less likely the bioplastic will undergo deformation when subjected to a force. The results in Figure 3 show that the young modulus was proportional to the tensile strength of the bioplastic but inversely proportional to elongation at break. The higher the concentration of sugar palm dreg and chitosan, the greater the value of the young modulus. It was because sugar palm dreg contained crude fiber, making the bioplastic stiff, while chitosan can bind the material molecules used to strengthen the bonds between them. According to Wiradipta (2017), adding cellulose can increase the tensile strength of plastic films. The increase in the tensile strength will be followed by an increase in the young modulus because tensile strength is proportional to the young modulus. As for glycerol, the modulus young was getting smaller along with the addition of glycerol.

Physical Properties

The physical characterization of bioplastics in this study included thickness and biodegradation percentage. The results of the measurement of the physical response parameter are shown in Table 3.

The results related to the bioplastics thickness in Figure 4 show that the higher concentration of the factors, the thicker bioplastics produced. It was because the thickness of the plastic is affected by the composition and properties of the materials used. These results are in accordance with previous studies conducted by Ibrahim *et al.* (2020), which stated that the higher concentration of sugar palm fiber (SPF) increases porosity formation and generates a heterogeneous surface, leading to thicker and more coarse films.

Biodegradation is the ability of bioplastics to be naturally decomposed by decomposer microorganisms. A high percentage of biodegradation indicates that bioplastics are degrading faster. The results in Figure 5 show that the

higher the concentration of sugar palm dreg and chitosan, the lower the percentage of biodegradation obtained. Because sugar palm dreg contained crude fiber, it was difficult to decompose by decomposer. Chitosan is hydrophobic, so it cannot absorb water from the soil and inhibits the degradation process. The percentage of biodegradation will increase with the addition of glycerol. According to Anita *et al.* (2013), glycerol has an important role in the biodegradation process because it has an (OH) group that can initiate a hydrolysis reaction after absorbing water from the soil so that the cellulose polymer will be decomposed into small pieces until it disappears in the soil. The polymer will be degraded due to the deterioration process or quality degradation because of breaking the chain bonds in the polymer.

Barrier Properties

The barrier characterization of bioplastics is measured as water vapor permeability (WVP), presented in Table 4.

Table 4. Barrier properties of the bioplastic

Experiment	Water Vapour Permeability (g.mm/kPa.hour.m ²)
A1B1C1	0.56±0.12
A1B2C2	0.43±0.04
A1B3C3	0.60±0.07
A2B1C2	0.77±0.15
A2B2C3	0.53±0.16
A2B3C1	0.34±0.04
A3B1C3	0.64±0.16
A3B2C1	0.70±0.16
A3B3C2	0.46±0.08

Note: (A) factors of sugar palm dreg, (B) factor of chitosan, (C) factor of glycerol. The numbers following the letters are the level factor ratio.

Table 5. Grey Relational Grade (GRG)

Experiment	Grey Relational Grade
A1B1C1	0.47
A1B2C2	0.53
A1B3C3	0.64
A2B1C2	0.57
A2B2C3	0.56
A2B3C1	0.58
A3B1C3	0.63
A3B2C1	0.70
A3B3C2	0.61

Note: (A) factors of sugar palm dreg, (B) factor of chitosan, (C) factor of glycerol. The numbers following the letters are the level factor ratio.

Water vapor permeability is the ability of plastics to allow water vapor to pass through it at a certain temperature and humidity. The lower the water vapor permeability, the better the plastic quality because it can defend against water vapor transfer to the packaging. The results in Figure 6 show that the higher concentration of sugar palm dreg and glycerol, the greater the water vapor permeability value. The increasing concentration of glycerol increased the moisture of the plastic because glycerol had hygroscopic properties, so it was incorporated between the plastic polymer chains (Hardjono *et al.*, 2016). Meanwhile, the higher the chitosan concentration decreased the water vapor permeability value. It was because chitosan is hydrophobic, inhibiting water vapor in the environment from penetrating or passing through the bioplastic. According to Giovanni *et al.* (2013) in Mustapa *et al.* (2017), chitosan has a hydroxyl group (OH) which is negatively charged, and an amine group (NH₂), which is positively charged so that chitosan can bond strongly ionically. The presence of a hydroxyl group with a negative charge in chitosan causes the chitosan to be hydrophobic, reducing the transfer rate of water vapor on the edible film.

Table 6. Response Table for Grey Relational Grade

Level	A	B	C
1	0.5487	0.5558	0.5833
2	0.5692	0.6008	0.5698
3	0.6471	0.6086	0.6120
Delta	0.0984	0.0527	0.0422
Rank	1	2	3

Note: (A) sugar palm dreg, (B) chitosan, (C) glycerol

Multiresponse Optimization

Multiresponse optimization aimed to find the optimal factor combination of all responses simultaneously. Merging multiresponse into a single response unit is carried out using the Gray Relational Analysis approach. The result of multiresponse merging is presented in Table 5.

These results were then analyzed using the Taguchi method to obtain the optimal combination of factors as presented in Table 6 and Figure 7. The optimal combination was obtained from the factor level with the largest response value. The ranking shows the most affected factors in the multiresponse.

Based on Table 6 and Figure 7, the optimal factor-level combination in the fabrication of sugar palm dreg bioplastic was A3B3C3 (consisting of 3 grams of palm sugar, 2 grams of chitosan, and 3 grams of glycerol). The factors that most affected multiresponse were sugar palm dreg-chitosan-glycerol, respectively. The characterization results show that the optimal bioplastic has a tensile strength of 1.46 MPa, 24.49% of elongation at break, 6.08 MPa of young modulus, 0.28 mm of thickness, 100% of biodegradation, and 0.61 g.mm/kPa.hour.m² water vapor permeability. These results show that the sugar palm dreg bioplastic was less resistant to mechanical loads but was elastic enough and had high biodegradation percentage.

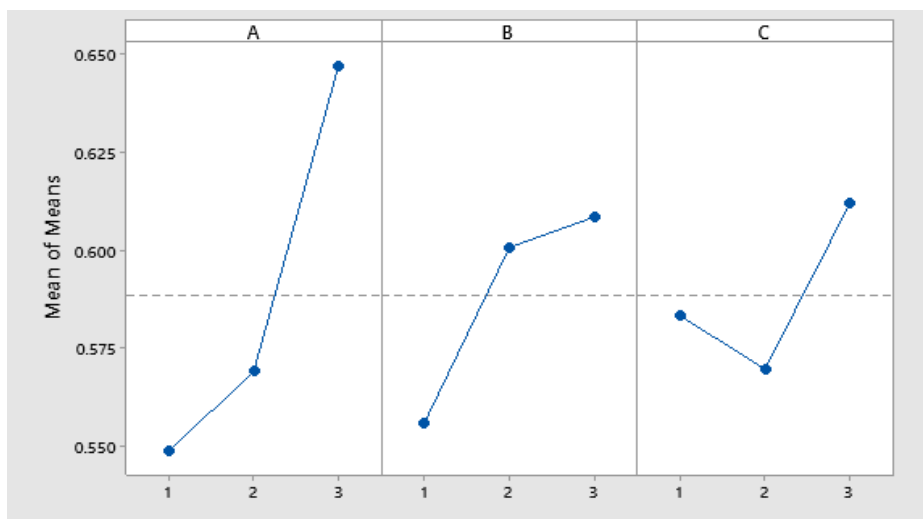


Figure 7. Main effect plot for Grey Relational Grade

CONCLUSION

Based on the research, it concluded that sugar palm dreg addition increased the tensile strength, young modulus, and bioplastic thickness but decreased the elongation at break. Adding chitosan affected the water vapor permeability, and glycerol increased the biodegradation percentage. The optimal combination of sugar palm dreg bioplastic was 3 grams of sugar palm dreg, 2 grams of chitosan, and 3 grams of glycerol. It had a tensile strength of 1.46 MPa, 24.49% of elongation at break, 6.08 MPa of young modulus, 0.28 mm of thickness, 100% of biodegradation, and 0.61 g.mm/kPa.hour.m². It was crucial to thoroughly evaluate and analyze the factors incorporated into the research to enhance this study. Additionally, it would be advantageous to identify a more practical approach to producing sugar palm flour.

From the result of this research, it can be concluded that microencapsulation of sweet potato leaf extract encapsulated by glucomannan and maltodextrin can be used as a natural source of antioxidants. The best treatment was the microencapsulation treatment of sweet potato leaf extract with a concentration of 10% maltodextrin combined with 0.75% glucomannan. In the best sample, it is known that the number of uniform particle size distributions is 0.296 μm (volume 78.3), and the Pdi value is 0.304 which indicates that the particle size is homogeneous. The results of phenolic dyes and SEM also showed that encapsulation of sweet potato leaf extract had occurred in the coatings (glucomannan and maltodextrin).

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