

Comparison of *Carboxymethyl Cellulose* (CMC) and *Microcrystalline Cellulose* (MCC) as Filler for Sago/Citric Based Bioplastic by Response Surface Methodology

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Abstract— Bioplastic material has emerged as one solution to the environmental problem caused by commercial plastic. Several raw materials have been used for bioplastic production, such as cassava, potato, and sago starch. This research focused on studying the effect of fillers and plasticizers variation to tensile strength and its morphology by using sago starch/citric acid-based bioplastic. The comparison of sorbitol and glycerol role as the plasticizer and *microcrystalline cellulose* and *carboxymethyl cellulose* as fillers were observed. This study was conducted by mixed and heated sago starch and fillers together. Then, citric acid and plasticizers were employed in the mixture solution. Next, the mixture was heated for 24 h then molded. The concentration of fillers, plasticizers, and citric acid were used as an independent variable which the interaction among them were investigated using response surface methodology (RSM) based on Central Composite Design (CCD). ASTM D822 evaluated tensile strength, and the morphological analysis was observed by using scanning electron microscopy (SEM). The results showed that the highest tensile strengths were 8.23 MPa for the glycerol and CMC process and 15.84 MPa for the sorbitol and MCC process. It was found that sorbitol and microcrystalline cellulose (MCC) increased the value of tensile strength twofold more than other fillers and plasticizers. As for the response surface method, the results describe the significant interaction between plasticizer and filler. It showed the tendency of increasing the concentration of fillers and citric acid decreased the tensile strength.

Keywords— Bioplastic; citric acid; filler; plasticizer; sago; tensile strength.

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I. INTRODUCTION

Plastics have been used for various life fields because it is considered economical, flexible, lightweight, and not easily broken material. This high consumption of plastics made Indonesia suspected as the second-largest country which contributed to plastic pollution, especially in the marine area [1]. It triggered many problems with the environment, not only in marine but also in soil and air. The plastics buried in landfills make the environment more severe because it is hard to degrade and diminish in the soil [2][3]. On the other hand, bioplastics are designed to be environmentally friendly since their carbon footprint is less than conventional plastic. Researchers also found that polysaccharides as a material source of bioplastic have been scrutinized for their biodegradable ability, and thermoplastic properties have become more fascinating in the material field [4]. One of the

bioplastics materials that are easily broken down by microorganisms is starch. In addition to its biodegradable nature, starch is a natural resource which renewable and has affordable prices, which makes starch is a promising material to be used as a matrix with reinforcements and fillers to form biopolymer [5] [6][7].

Sago (*Metroxylon spp*) has predominant starch, which grows in freshwater swamp areas, peat swamps, areas along rivers, areas around water sources, or swamp forests. Indonesia is one of the broadest sago plantations globally that spread out from West Papua to Aceh and Riau Province [8]. Sago has a similar physicochemical of starch like cassava and potato. Amylopectin content in the sago starch positively affects tensile strength, which makes this material suitable as bioplastic's source [9].

One application of bioplastics is edible film. However, this starch-based edible film also has some disadvantages, like

weak mechanical and barrier properties. Besides, it has high water vapor permeability and high brittleness, which undesirable for packaging used. Therefore, plasticizing agents and filler help to minimize this condition [10], [11]. Bioplastics or biodegradable plastic still have some disadvantages compared to conventional plastics; those are mechanical properties that are easily torn and not as strong as conventional plastic. Some research has been carried out to develop sago starch-based bioplastics' ability to have superior properties by combining starch with other polymers.

Ghanbarzadeh *et al.* [12] studied the physical properties of edible films with modified starch-carboxymethyl cellulose (CMC). The best results in this study were 15% w/w of carboxymethyl cellulose (CMC)/starch. It caused decreasing in moisture absorption and film solubility from 24.34% and 23.76% to 20.00% and 19.89%, respectively. Zuraida *et al.* [13] also reported a study with the effect of water and citric acid on sago starch-based bioplastics. They varied glycerol and citric acid concentration and weight percentage of water-based on 100%(w/w) sago starch. The results showed that the optimum tensile strength for the composition of sago and glycerol was 23.06 MPa, the composition of sago starch-glycerol and citric acid was 24.766 MPa. Aquades as an additive in sago starch-glycerol has an optimum tensile strength of 3.29 MPa.

Wilpiszewska and Czech [14] conducted a modification of citric-starch potatoes with microcrystalline cellulose (MCC) amplifiers with variations of MCC (1, 3, 5, 7, 10, and 15%w/w). The results exhibited that the addition of microcrystalline cellulose (MCC) increased the polysaccharide film's mechanical properties. The young's modulus and tensile strength increased from 42 to 96 MPa and 2.6 to 3.9 MPa, respectively for non-filled starches or citric acid films and the same system containing 15%w/w MCC. In addition, cellulose fillers' presence influencing the thermal properties of starch-based materials by increasing the transition MCC glass content increased from 108 to 129 °C, each for non-filled starch or citric acid films and the same system having 15%w/w MCC. Also, starch-based films' hydrophilicity properties are reduced because the absorption of moisture and solubility in water decreased.

Another study investigated the concentration of plasticizer on mechanical and barrier properties of sweet potato starch film. The high concentration of plasticizers such as glycerol and sorbitol can reach high water solubility and elongation percentage. The contrary results showed that plasticizer's increased concentration could decrease the puncture strength value [15].

Citric acid has been used as a cross-linking agent, which non-toxic, low cost, and able to stabilize the polysaccharide material. Several studies about the ability of citric acid as a cross-linking agent have been reported [14], [16], [17]. Priyadarshi *et al.* [16] reported that citric acid employment decreased moisture content and water absorption and generated the water vapor barrier. Meanwhile, another study mentioned the less concentrated citric acid produced a good cross-linking agent for polysaccharides [17]. Besides, Ma *et al.* [18] mentioned that citric acid concentration has also influenced edible film's mechanical properties. Based on previous research, which generally developing the mechanical and properties of bioplastics, varying composition

of raw materials, plasticizers and fillers were used. However, the mechanical, morphological, and biodegradability characteristics produced are not consistent with the different types and compositions of plasticizers, fillers, and raw materials used. Therefore, this study aimed to investigate the effect of glycerol and sorbitol as a plasticizer and microcrystalline cellulose and carboxymethyl cellulose as fillers for sago-based bioplastic. Response surface method was applied to evaluate the interaction between plasticizers and fillers towards tensile strength.

II. MATERIAL AND METHOD

A. Material and Apparatus Tools

The raw material used in this study was sago starch originating from the Selat Panjang region, Meranti Islands. The fillers were microcrystalline cellulose (MCC) and carboxymethyl cellulose (CMC), sorbitol, and glycerol as a plasticizer were purchased from Brataco Chemika Inc. (Indonesia), and citric acid was purchased from Putra Riau Bersama (PRB, Indonesia) and aquadest. The equipment was analytical balance, beaker, volumetric flask, goblet glass, measuring cup, stirring rod, oven, filter, glass mold, watch glass, heating mantle, condenser, triple neck flask, thermometer, and hot plate.

B. Methods

The procedures are divided into two parts. There is the preparation of filler solutions and bioplastic mold. The brief procedure is shown in Fig. 1.

1) *Preparation of Microcrystalline Cellulose and Carboxymethyl Cellulose Solution:* MCC with concentrations 15, 20, and 25 % (w/w starch) was placed with 100g water in a flask and mixed for 30 min using a mechanical stirrer at 90° C until a homogeneous MCC solution was formed. Meanwhile, for the CMC, it is homogenized with 100 ml equates at 75°C, 100 rpm for 15 min.

2) *Preparation of Bioplastic:* Dissolved 10g of sago starch with 100 ml of distilled water in beaker glass at room temperature (25 °C). Then, added the MCC solution. The mixture was stirred around 200 rpm and heated up at 90 °C (kept it constant) after 20 min sorbitol was employed. Citric acid was added at 25th min. The total process was conducted for 30 minutes. The homogeneous solution was stand for ± 5 min, then it was poured into the mold for 24 hours. Once it dried, the plastic film was released from the mold by lifting a thin sheet from one side of the horizontal direction slowly until the entire bioplastic surface was detached from the mold. Afterward, bioplastics were tested for characteristic and mechanical properties—repeated procedures for variations of citric acid levels, sorbitol plasticizer levels, and MCC filler levels. Also, for CMC and glycerol variations.

C. Bioplastic Characteristics Test

The characteristic tests were conducted to analyze the mechanical and morphological properties.

1) *Mechanical Properties:* Characterization of the mechanical properties of bioplastics included tensile strength, modulus young, and elongation. This mechanical property

test refers to the ASTM D822 standard with 2 x 6 cm sample size test specimens.

2) *Morphological*: The structure of bioplastic morphology was analyzed by using scanning electron microscopy (SEM) which refers to the JSM-6390A standard. This SEM analysis aims to see the phase spread of the bioplastic sample surface and examined the aggregation of cellulose fillers in their matrix.

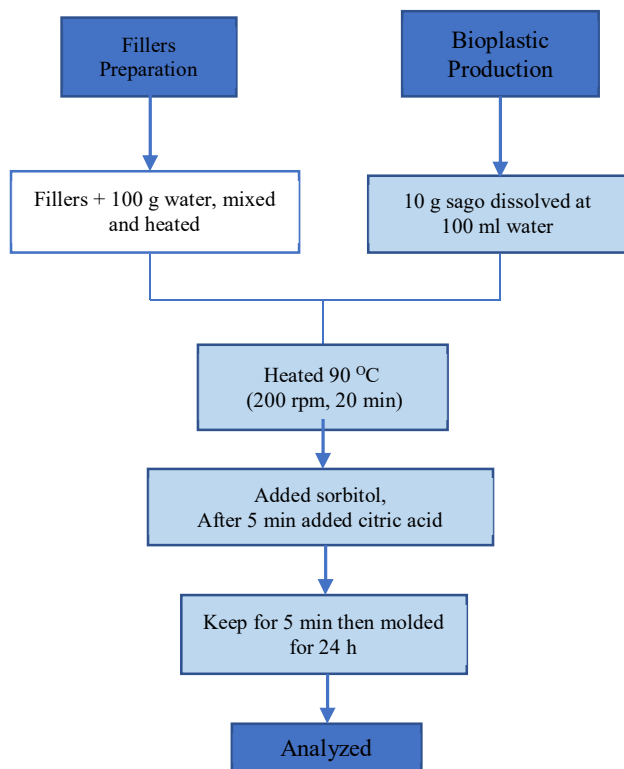


Fig. 1 Flowchart procedure of sago-based bioplastic.

D. Experimental design

Central composite design (CCD) was employed in this research to model the equations. Percentage of citric acid (X_1), plasticizer (X_2), and filler (X_3) were varied to describe the correlation among the parameters. They were the independent variables to figure out the earlier research related to bioplastic from sago starch. Tensile strength stands for response was dependent variable Y_i , while the concentration of citric acid (x_1), plasticizer (x_2), and filler (x_3) are listed in Table 1. The experiment was conducted for twenty runs at three levels with the axial point on two responses (tensile strength for glycerol & CMC (Y_1) and sorbitol and MCC (Y_2)). The experimental data are given in Table 2. The relation among the parameters was described by using Response Surface Method (RSM).

TABLE I
THE RANGE AND LEVEL OF INDEPENDENT VARIABLES FOR BIOPLASTIC PRODUCTION

Level code	Citric Acid (x_1)	Plasticizer (x_2)	Filler (x_3)
-1	3	25	15
0	6	30	20
1	9	35	25

TABLE II
EXPERIMENTAL RESULT FOR BIOPLASTIC PRODUCT WHICH VARIED PLASTICIZER AND FILLER

No.	Citric acid (%), X_1	Plasticizer (%), X_2	Filler (%), X_3	Tensile strength (MPa), Y_1	Tensile strength (MPa), Y_2
1	3	25	15	6.24	9.31
2	9	25	15	5.91	8.2
3	3	35	15	5.59	6.3
4	9	35	15	4.8	4.25
5	3	25	25	8	10.26
6	9	25	25	8.07	9.83
7	3	35	25	7.09	8.49
8	9	35	25	6.14	7.51
9	0.955	30	20	7.68	15.84
10	11.045	30	20	6.01	11.04
11	6	21.59	20	6.3	10.13
12	6	38.409	20	4.77	4.9
13	6	30	11.59	5.65	5.13
14	6	30	28.41	8.23	9.34
15	6	30	20	5.16	13.26
16	6	30	20	6.7	12.43
17	6	30	20	6.21	10.75
18	6	30	20	5.75	14.34
19	6	30	20	6.04	11.46
20	6	30	20	7.02	11.69

* Y_1 showed the result for a condition using glycerol as plasticizer and CMC as filler; on the contrary, Y_2 for sorbitol as plasticizer and MCC as filler.

Quadratic equation produced empirical models for tensile strength for varied plasticizer and filler as shown in Equation 1.

$$Y_i = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \quad (1)$$

where Y_i is response function, β_0 is intercept term, $\beta_1, \beta_2, \beta_3$ are linear effect, $\beta_{11}, \beta_{22}, \beta_{33}$ are quadratics effect and $\beta_{12}, \beta_{23}, \beta_{13}$ are interaction effect among the variables. P-value was applied to relate the effects of the variables with responses and the F-value was checked the validity of estimated model equation for tensile strength. RSM was analyzed by using STATISTICA version 7 program.

III. RESULT AND DISCUSSION

A. Bioplastic Morphology

The samples were assessed using Scanning Electron Microscope (SEM) to observe the surface structure and the filler's distribution on starch, as shown in Fig. 2. It is represented by 30% plasticizer, 28.41% of CMC filler, and 6% citric acid. Fig. 2a shows that CMC as the filler has not fully distributed on the surface, and CMC agglomerated in certain edible film spots. It is caused by the increase of filler content in the samples. The increasing agglomeration made the interaction between fillers and matrix weak [19]. On the other hand, the distribution of MCC as filler is shown in Fig. 2b. It was represented by 20% filler of MCC, 30% sorbitol as a plasticizer, and 9.5% citric acid. SEM shows a rough structure and an untidy but rigid one.

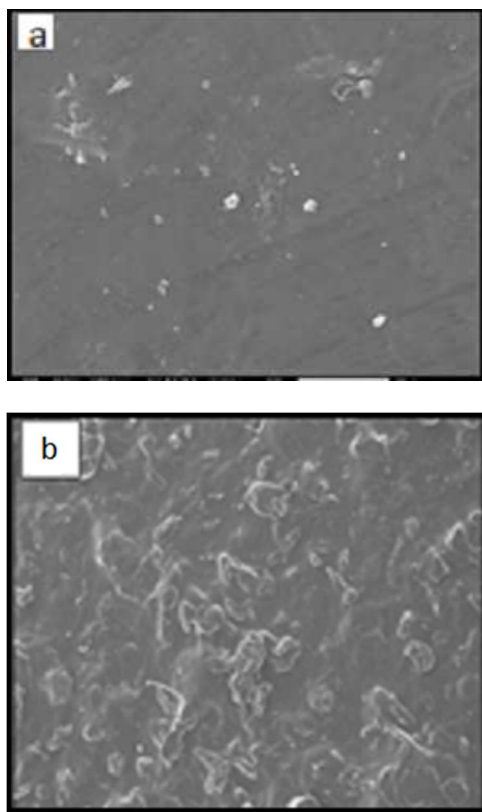


Fig. 2 SEM's result used (a) CMC and (b) MCC as filler for magnified 500 μm

Since sorbitol has a more significant molecule, the matrix is occupied with hydrogen bonding, which disrupts polymer structure into an untidy structure [15], [20]. Agglomeration occurred in the matrix initiated the decreasing of tensile strength. It made the filler unable to support the tensile transfer evenly. It triggered unstable conditions for the enhancement filler mechanism [19].

B. Experimental Design and Statistical Analysis

New model equations were produced from the experimental data laboratory to predict the tensile strength by using a statistical program. An ANOVA system has been employed to correlate the relation among the parameters [21]. The model equation for tensile strength by using glycerol as plasticizer and CMC as filler (Y_1) and sorbitol as plasticizer and MCC as filler (Y_2) are given in Equations 2 and 3, respectively.

$$\begin{aligned}
 y_1 = & -0.5171 - 0.1229X_1 + 0.5847X_2 \\
 & - 0.1436X_3 + 0.01279X_1^2 \\
 & - 0.0085X_2^2 + 0.0114X_3^2 \\
 & - 0.0123X_1X_2 + 0.002X_1X_3 \\
 & - 0.0054X_2X_3
 \end{aligned} \quad (2)$$

$$\begin{aligned}
 Y_2 = & -75.9179 - 0.4169X_1 + 4.1832X_2 \\
 & + 2.9854X_3 + 0.0158X_1^2 \\
 & - 0.0781X_2^2 - 0.0821X_3^2 \\
 & - 0.0124X_1X_2 + 0.0146X_1X_3 \\
 & + 0.0143X_2X_3
 \end{aligned} \quad (3)$$

Regression analysis for tensile strength is depicted in Table 2. Analysis of variance (ANOVA) was used with a 5% level of significance, and the model was set up by checking the F-

value. The negative sign in the front term indicated the antagonist effect of the variables [22]. Table 3 shows that F value was higher than the F table, meaning the variables significantly affected tensile strength and rejected the null hypothesis.

The coefficient of correlation, R^2 , was accepted if it higher than 0.75, which implied the relation between the experimental and predicted value of tensile strength. Figure 3 illustrates the predicted and experimental value of tensile strength for Y_1 , which uses glycerol as plasticizer and CMC as filler, and Y_2 , sorbitol as a plasticizer, and MCC as filler. It showed the close agreement between the observed and predicted value and yield R^2 was 0.875 (Y_1) and 0.913. It indicated that the proposed equation was fitted to experimental data.

TABLE III
ANOVA FOR TENSILE STRENGTH

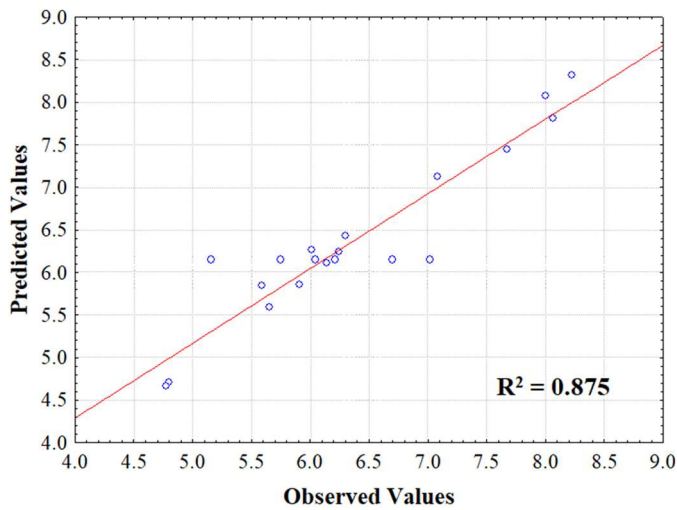
Mode	Sum of Squares	df	Mean Square	F value	F 0.05
Tensile strength (Y_1)					
Regression	17.7976	9	1.9775		
Residual	2.5253	10	0.2525	7.8308	3.02
Total	20.3229	19			
Tensile strength (Y_2)					
Regression	167.8598	9	18.6511		
Residual	15.9357	10	1.5935	11.7039	3.02
Total	183.7956	19			

TABLE IV
THE SIGNIFICANCE OF COEFFICIENT ESTIMATED FOR TENSILE STRENGTH (Y_1)

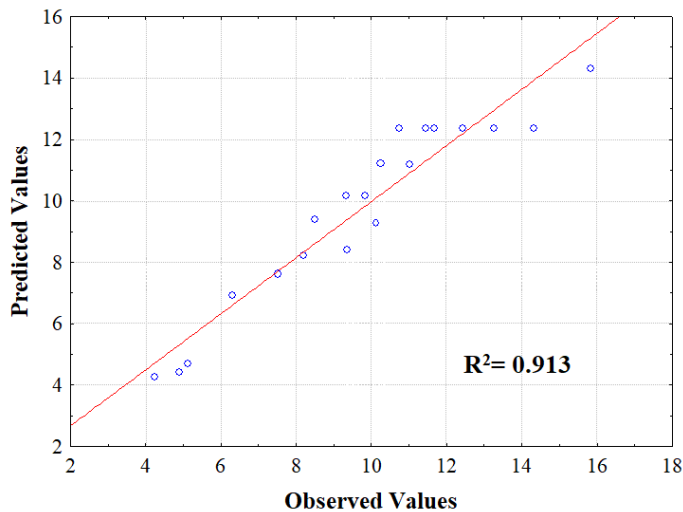
Source	Effect	Sum of Squares	df	Mean Square	F value	p-value
X_1	-0.7042	1.6931	1	1.69307	6.7044	0.02698
X_1^2	0.5033	0.9122	1	0.91224	3.61244	0.08653
X_2	-1.0504	3.7679	1	3.76785	14.9204	0.00315
X_2^2	-0.4231	0.6452	1	0.64516	2.5548	0.14104
X_3	1.6253	9.0202	1	9.02021	35.7194	0.00014
X_3^2	0.5702	1.1719	1	1.17188	4.6405	0.05666
X_1X_2	-0.3700	0.2738	1	0.27380	1.0842	0.3223
X_1X_3	0.0600	0.0072	1	0.00720	0.0285	0.8693
X_2X_3	-0.2700	0.1458	1	0.14580	0.5774	0.4649

Table 4 and 5 illustrate the significant variable among the variables for two responses (Y_1 and Y_2). The significance of coefficients for Y_1 depicted that concentration of citric acid, plasticizer, and filler has a significant effect on tensile strength among other variables. It also found that there was no significant influence for interaction between variables and

the quadratic mode. The higher F-value was concentration filler, and it fitted for p-value.



(a)



(b)

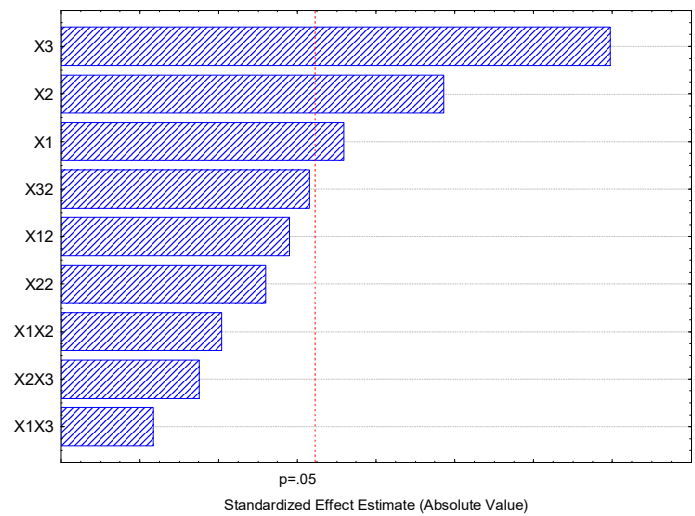
Fig. 3 Observed vs predicted tensile strength in bioplastic product (a) for response Y_1 , (b) response Y_2 .

Table 5 shows x_1 , x_2 , x_2^2 , x_3 , x_3^2 acted as significant variables for Y_2 . It had expected that F value and P value had a significant effect on x_1 , x_2 , x_2^2 , x_3 , x_3^2 which named for concentration of citric acid, plasticizer linear and quadratic, filler in linear and quadratic. Filler concentration has higher F-value than other variables and it fitted the p-value.

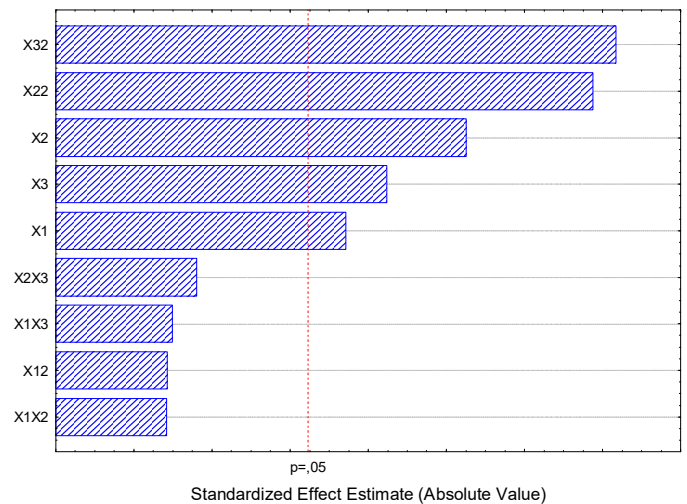
Student t-distribution was applied to estimate the value of equation parameter and error standard to see the confidence limit, while p value was employed to check the significance of variables for tensile strength properties. Both tests were used to seek the significant effect for each variable in the process; this correlation is illustrated in Fig. 4. Fig. 4 (a) depicts that the significant concentration of filler, plasticizer and citric acid with $p > 0.05$, on the other hands, Fig. 4(b) illustrates the interaction of quadratic filler, plasticizer and linear variable of plasticizer, filler and citric acid which have significance influence on tensile strength.

TABLE V
THE SIGNIFICANCE OF COEFFICIENT ESTIMATED FOR TENSILE STRENGTH (Y_2)

Source	Effect	Sum of Squares	df	Mean Square	F value	p-value
X_1	-1.8515	11.7033	1	11.7033	7.34406	0.021934
X_1^2	0.2838	0.2901	1	0.2901	0.18204	0.678662
X_2	-2.9066	28.8463	1	28.8463	18.10164	0.001677
X_2^2	-3.9053	54.9591	1	54.9591	34.48798	0.000157
X_3	2.2128	16.7188	1	16.7188	10.49137	0.008884
X_3^2	-4.1026	60.6652	1	60.6652	38.06866	0.000106
X_1X_2	-0.3725	0.2775	1	0.2775	0.17414	0.685272
X_1X_3	0.4375	0.3828	1	0.3828	0.24022	0.634620
X_2X_3	0.7175	1.0296	1	1.0296	0.64610	0.440198



(a)



(b)

Fig. 4 Pareto chart of response for Y_1 (a) and Y_2 (b)

The higher significance effect is shown by quadratic filler in Figure 4. The lowest p-value ($p < 0.05$) indicated the interaction between citric acid and filler (Fig. 4a) and interaction between citric acid and plasticizer for tensile strength properties.

C. Interaction of Variables for Tensile Strength Properties

All the three-dimensional responses surface is illustrated in Fig. 5 and 6. The interactive effect of each variables with respect to tensile strength were analyzed. Fig. 5 shows increased plasticizer has significance effect and influence the tensile strength than increased citric acid. The lowest tensile strength was achieved at 3% citric acid, 15% filler and 35% plasticizer about 5.59 MPa.

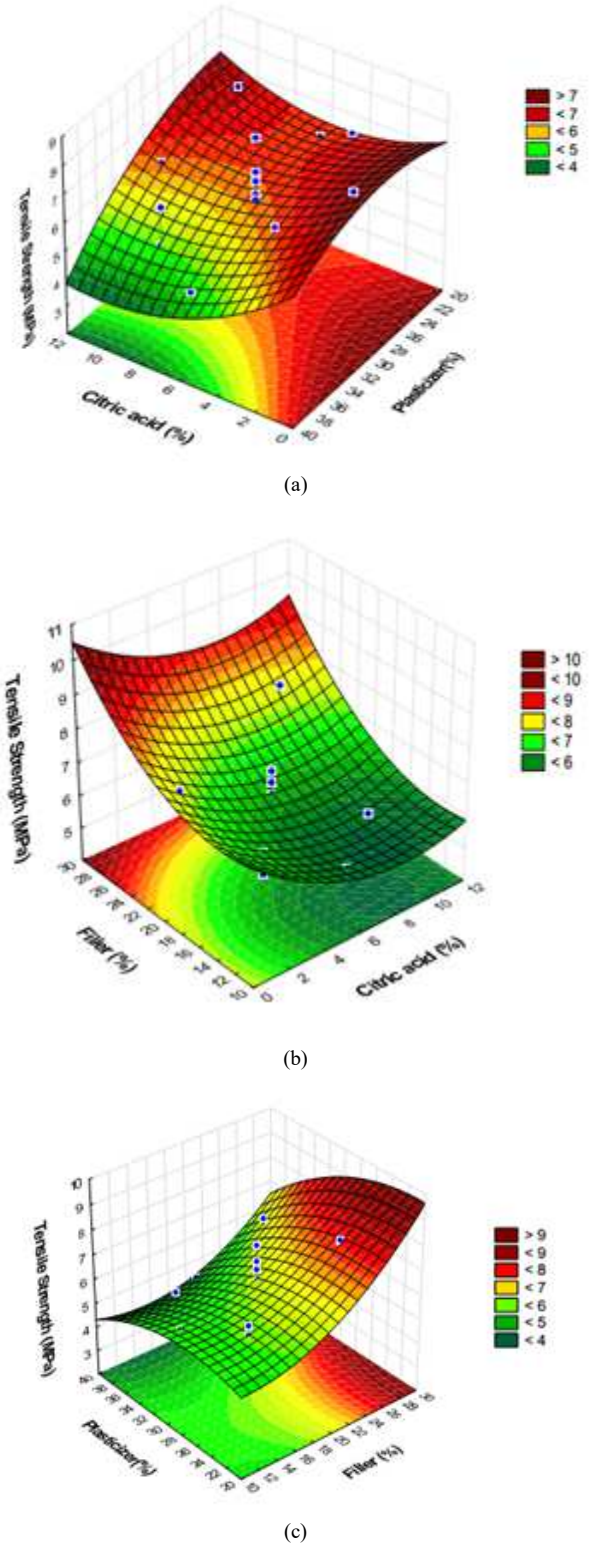


Fig. 5 Response fitted surface area of tensile strength for Y₁ (a) citric acid and plasticizer, (b) citric acid and filler (c) filler and plasticizer.

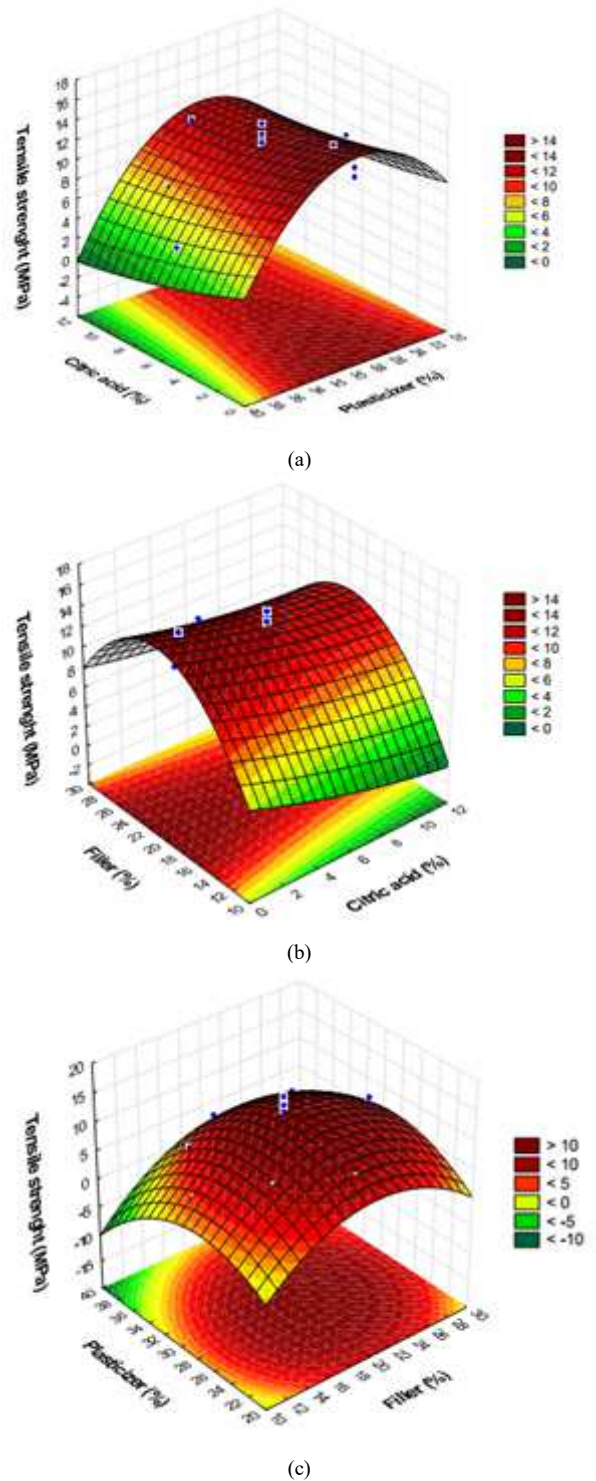


Fig. 6 Response fitted surface area of tensile strength, Y₂, (a) Citric acid and plasticizer (b) citric acid and filler (c) Filler and plasticizer.

The tensile strength value was about 4.77-8.23 MPa. Fig. 5 shows that the concentration of filler and plasticizer has a significant effect on increased tensile strength. According to [23], CMC interacted and substituted the hydrogen bond generated a dense blending product structure. The intensive bonding between starch and CMC made deprotonation of CMC's carboxyl group and stretching of carbonyl (-CO) group presence. This condition enhanced the mechanical properties of the edible film, including tensile strength. Another study [24] found that glycerol made a polymorphic

transition that shrinks the starch's crystallites. This hydroxyl group caused the increased film strength. The high concentration of plasticizer (in the case of; glycerol) decreased the molecule's surface tension that arranged matrix film, thus decreasing edible film [25]. Fig. 5 (a and b) illustrates that the concentration of citric acid has not significantly affected tensile strength.

From Fig. 6 (a, b and c) illustrates that the concentration of plasticizer and filler has a significant effect on tensile strength. It was also reported by another studies [15], [23] where they mentioned glycerol and sorbitol contributed through hydrogen bonding to make a strong interaction bonding between polymer and plasticizer. Moreover, Martinez *et al.*, [15] reported that the increasing concentration of sorbitol or glycerol can decrease the puncture strength since the plasticizer affects the movement of the polymer chains.

The dome graphs display high tensile strength in higher plasticizers (in terms of sorbitol) and filler (MCC). Maximum tensile strength was achieved at 30% plasticizer and 20% filler for 15.84 MPa in the dome's peak whereas from the center point. The citric acid (3-9%) depicts that the higher concentration of citric acid has not significantly affected tensile strength.

IV. CONCLUSIONS

The model equation for tensile strength by using glycerol as plasticizer and MCC as filler (Y_1) and sorbitol as plasticizer and MCC as a filler (Y_2) are given in Equations 2 and 3, respectively. It was found that citric acid as cross-linking agent has higher tensile strength than without citric acid and that sorbitol and microcrystalline cellulose (MCC) has higher tensile strength than glycerol and carboxymethylcellulose (CMC). Also, it showed that plasticizer concentration had a significant effect on tensile strength. The best edible film composition consists of microcrystalline cellulose (MCC) 20% w/w, sorbitol 30% w/w with citric acid 0.95% w/w. The best tensile strength value was 15.84 MPa.

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