

## Physical, Physicochemical, Mechanical, and Sensory Properties of Bioplastics from Phosphate Acetylated Arenga Starches

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Bioplastics are alternative to plastic packaging made from renewable natural materials. They have a great potential for wider application due to their environmental-friendliness and ease of degradation. This research, therefore, aimed to evaluate the physical, physicochemical, mechanical, and sensory characteristics of bioplastics made from native arenga starch (NAS) and phosphate acetylated arenga starch (PAAS). The PAAS was obtained by dual modification of NAS through acetylation using 5% acetic anhydride and crosslinking using a mixture of sodium trimetaphosphate (STMP) and sodium tripolyphosphate (STPP) at 99:1 (*w/w*). The concentrations of the mixture were varied at 2, 4, 6, 8, 10, and 12% (*w/w*) of the starch. The thickness, water holding capacity (WHC), oil holding capacity (OHC), water vapor transmission rate (WVTR), water content, biodegradation, Fourier transform infrared (FT-IR) spectroscopy, tensile strength, elongation at break, Young's modulus, and sensory properties of the NAS and PAAS bioplastics were investigated. The results showed the thickness of the NAS and PAAS was generally uniform. The WHC of the NAS bioplastic was higher than that of PAAS. The OHC and WVTR of the PAAS bioplastics increased with the increment in the concentration of the STMP/STPP mixture. Furthermore, the water content of the PAAS bioplastics was lower than that of NAS, while the weight loss due to biodegradation of the NAS was higher compared to PAAS. The PAAS bioplastics were characterized by FTIR, which confirmed the acetylation and crosslinking between the arenga starch molecules. Generally, the elongation at break of the PAAS bioplastics was higher than that of the NAS bioplastic, color of the PAAS bioplastics was more transparent and texture of the PAAS bioplastics surface was smoother than of the NAS bioplastic.

### INTRODUCTION

Packaging is the final part of the processes involved in the production of food and non-food products purposely to increase shelf life, improve consumer acceptance, and as a means of providing support for the distribution and expansion of product marketing [Prasteen *et al.*, 2018]. The major material used for this purpose is plastic and this has led to a continuous increase in its waste thereby causing environmental pollution because it is non-biodegradable, its recycling process is quite expensive, and it has the ability to contaminate foods [Ogunrinola & Akpan, 2018]. There is, however, the urgent need to find alternative materials for packaging and one of these is the bioplastics due to its safe and biodegradable nature.

Bioplastics are produced from renewable natural materials and have been observed to have the potentials of being

an alternative to plastic packaging due to their environmental friendliness and easy degradation. Therefore, there is an urgent need for bioplastics due to the rapid increase in the pace of plastic production and air pollution causing several health risks because of their toxic nature [Jain & Tiwari, 2015]. Bioplastics are, however, very suitable alternatives to improve the quality of life and maintain a pollution-free planet [Keziah *et al.*, 2018].

It has been discovered that bioplastics made from native starch have poor physicochemical, mechanical, functional and sensory characteristics compared to those modified chemically. According to Shindu & Khatkar [2018], those produced from modified wheat starch are transparent, have greater tensile strength, and lower solubility compared to the native wheat starch. Moreover, edible films of acetylated rice starch were also found to be stable to heat, have high elongation at break, and a rapid process of degradation [Colusi *et al.*, 2017]. Meanwhile, the mechanical properties of bioplastics from tapioca starch modified using acetic anhydride were observed to be superior over these of the native starch

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[Tawakaltu *et al.*, 2015]. The elongation at break, solubility, and oxygen permeability characteristics of biodegradable films from hydroxypropyl sago starch were higher and the water vapor transmission rate was lower compared to the native material [Polnaya *et al.*, 2013].

One of the potential starches to be used as a bioplastic polymer was native arenga starch (NAS), but it is limited by fast retrogradation, inability to withstand acidic conditions, low viscosity, and paste resistance [Rahim *et al.*, 2019]. Therefore, there is a need for its modification in order to obtain its suitable properties required by bioplastic polymer materials. The novelty of this research was the use of dual modification of the main raw material through acetylation and crosslinking. This involved the development of a phosphate acetylated arenga starch (PAAS) through acetylation and crosslinking. These were intended to produce a modified material that is difficult to retrograde, stable to heat and acids, and resistant to water due to the presence of acetyl groups and crosslinking in the starch molecules. This was possible because previous studies have reported the incorporation of acetyl and phosphate in starch molecules was able to inhibit the retrogradation process and aid resistance to heat, acid-base, and water [Rahim *et al.*, 2017]. Therefore, the objective of this research was to evaluate the physical, physicochemical, mechanical, and sensory characteristics of the NAS and PAAS bioplastics obtained through acetylation using 5% acetic anhydride and crosslinked using a mixture of sodium trimetaphosphate (STMP) and sodium tripolyphosphate (STPP) at 99:1 (*w/w*) with different concentrations.

## MATERIALS AND METHODS

### Materials

The materials used in the research consisted of NAS extracted from the pith of sugar palm trees (*Arenga pinnata*). Distilled water and 98% acetic anhydride were purchased from Sigma-Aldrich (Steinheim, Germany). Hydrochloric acid (HCl), sodium hydroxide (NaOH), STMP, STPP, 96% ethanol, glycerol, and acetic acid were purchased from Merck (Darmstadt, Germany). All the chemicals used for the bioplastic analysis were of analytical grade.

### Arenga starch extraction

The NAS was prepared according to the method described by Sahari *et al.* [2014] with some alterations. *Arenga pinnata* tree was cut down and the stem was then split to release the woody fiber together with the starch from the inward delicate center of its stem. Step by step, water was brought inside the fiber and starch blend got from the stem and was completely massaged by hand. The blend was then separated to permit the water to move through the sifter with starch granules in the mixture. The starch was conceded sufficient opportunity to settle at the base of the compartment, and water was tapped. After that, the starch was kept in an outdoors for momentarily and then dried in the oven for 72 h at temperature 70°C.

### Preparation of dual modification arenga starch

Acetylation of NAS was conducted according to the method of Rahim *et al.* [2015] while crosslinking followed the Koo

*et al.* [2010] approach with a slight modification. The suspension consisting of 100 g arenga starch and 225 mL distilled water was stirred using a magnetic stirrer for 1 h at room temperature. This was followed by the dropwise of acetic anhydride in such a quality as to give 5% of weight of arenga starch (*w/w*). The pH of the suspension was maintained at 8.0 to 8.5 by adding 3% NaOH. After 60 min, the pH was increased to 10.5 through the addition of 3% NaOH. Next, a mixture of STMP/STPP at 99:1 (*w/w*) was added in such an amount to obtain concentration of 2, 4, 6, 8, 10, and 12% by weight of arenga starch (*w/w*). The suspension was again stirred for 30 min at room temperature and next 0.5 N HCl was added until the pH decreased to 4.5 to end the reaction. After reaction, the suspension was left to sediment for 30 min and the sediment was washed with distilled water three times and 96% ethanol once. Next, the sediment was dried in an oven at 60°C for 15 h, mashed, and filtered with 100 mesh sieve. The modified arenga starches were called 2, 4, 6, 8, 10, and 12% PAAS, respectively, and were used as bioplastic polymer materials.

### Preparation of bioplastics

Bioplastic was prepared according to the method developed by Chung *et al.* [2010] with a slight modification. The NAS and PAAS (2, 4, 6, 8, 10, and 12%) were weighed to be 10 g, and 150 mL of distilled water was added. The solutions were heated on a hot plate to a temperature of 100°C with stirring after which 10 mL of acetic acid and 3 mL of glycerol were added. The heating process continued until the gel started forming. The solution was further stirred for 10 min and was poured into stainless steel strips and dried at room temperature for 4 days. The bioplastics obtained were analyzed for physical, physicochemical, mechanical, and sensory properties.

### Thickness determination

Bioplastic thickness was measured according to the method proposed by Turhan & Sabhaz [2004] through the use of calipers. The samples were placed between the jaws of the caliper and the thickness was measured at five different places and the average was calculated.

### Water and oil holding capacity determination

The water and oil holding capacity (WHC and OHC, respectively) were determined using a method developed by Larrauri *et al.* [1996] with some modification. Briefly, 25 mL of distilled water or olive oil were added to 250 mg of bioplastic samples, stirred, and the mixture was left at room temperature for 1 h. After centrifugation at 3,500×g for 30 min (GS 150 Centrifuge, Clements, Sydney, Australia), the residue was weighed and the holding capacity of water and oil was calculated as g of water or oil, respectively per g of dry bioplastic sample.

### Water vapor transmission rate determination

Water vapor transmission rate (WVTR) was determined using the method proposed by Xu *et al.* [2004]. This involved the production of a saturated salt solution in a chamber using a jar with a diameter of 12 cm and a height of 15 cm as well as

the adjustment of the relative humidity (RH) in the chamber to 75% by adding a solution of 40% NaCl (*w/v*) at room temperature. Furthermore, an acrylic cup with diameter of 5 cm and height of 1.8 cm was filled with 10 g silica gel, covered with bioplastic according to its size, and placed in the 75% RH chamber. The water vapor diffusing through the bioplastics and silica gel added to the weight and the whole setup was weighed every hour for 8 h to determine the weight gain. The data was used to plot a graph of the time *vs.* weight, and the slope was recorded to calculate WVTR using the following equation:

$$\text{WVTR (g/h/m}^2\text{)} = \frac{\text{Change of bioplastics sample weight (g/h)}}{\text{Surface area of the bioplastics sample (m}^2\text{)}}$$

### Moisture content determination

Moisture content was measured following the method applied by AOAC [2005]. The portions of bioplastics (0.5 g) were dried in an oven at 105°C for 2 h.

### Biodegradability analysis

The biodegradability of the bioplastic samples was investigated according to the method developed by Ashok *et al.* [2018] with slight modification. The samples were cut into 1 × 4 cm, weighed ( $W_1$ ), and buried in the soil at a depth of 8 cm for a curing duration of 9 days after which they were weighed again ( $W_2$ ). The weight loss due to the biodegradation process was determined through the use of the following formula:

$$\text{Weight loss (\%)} = \frac{W_1 - W_2}{W_1} \times 100\%$$

### FTIR spectra analysis

FTIR spectra of NAS and PAAS bioplastics were analyzed according to Diop *et al.* [2011]. The bioplastics were mixed with KBr at a ratio of 1:100 (sample: KBr, *w/w*). The infrared spectra were obtained using a Fourier Transform Spectrometer (IR Prestige-21, Shimadzu Corporation, Tokyo, Japan) at a wavenumber range from 4000  $\text{cm}^{-1}$  to 500  $\text{cm}^{-1}$  regions.

### Tensile strength, elongation at break, and Young's modulus determination

Tensile strength and elongation at break were measured using a Mechanical Universal Testing Machine (Model LR30K, LLOYD Instruments Ltd., Hampshire, England). This involved cutting the bioplastics into the required standard and the two ends were clamped to the testing machine at a distance of 50 mm and a speed of 10 mm/min. The start knob was turned on and the machine pulled the sample until it broke and the tensile strength and elongation at break values were recorded afterward. The tensile strength was calculated based on the maximum force (N) applied for the material to break divided by the area of the bioplastic ( $\text{mm}^2$ ):

$$\text{Tensile strength (MPa)} = \frac{F_{\text{Max}} \text{ (N)}}{\text{Bioplastics surface area (m}^2\text{)}}$$

The elongation at break was calculated using the equation:

$$\text{Elongation at break (\%)} = \frac{\text{Maximum length} - \text{Initial length}}{\text{Initial length}} \times 100\%$$

Young's modulus was calculated based on the values of tensile strength and elongation at break according to the formula:

$$\text{Young's modulus (MPa)} = \frac{\text{Tensile strength}}{\text{Elongation at break}/100}$$

### Sensory evaluation

The sensory analysis was conducted using a 15-member panel consisting of students from the Faculty of Agriculture, Tadulako University, Central Sulawesi Indonesia. The bioplastic samples were cut into small pieces and the color, texture, aroma, and overall acceptability were tested using a 7-point hedonic scale where 7 indicates highly very like, 6 – very like, 5 – like, 4 – neither like nor dislike, 3 – somewhat like, 2 – dislike, and 1 – very dislike. The panelists were instructed to rate the attributes indicating their degree of likeness using the numbers provided in the hedonic scale according to their preference.

### Statistical analysis

All parameters were determined in triplicate except for thickness and sensory analysis, which were carried out four times. The results were analyzed through the application of one-way ANOVA using SPSS version 22 (SPSS Inc., Chicago, IL, USA). The means were compared with Duncan's multiple tests and the statistical significance was defined at  $p \leq 0.05$ .

## RESULTS AND DISCUSSION

### Thickness

The thickness of bioplastics of NAS and PAAS obtained using different concentrations of the STMP/STPP mixture are shown in Figure 1. There were no significant differences ( $p > 0.05$ ) in the thickness of both NAS and PAAS bioplastics and this was associated with the equal amount of raw material used. The values (0.36–0.45 mm) were in line with the findings of Marichelvam *et al.* [2019] that the average thickness of bioplastics made from corn and rice starch was 0.25 mm and this means they can be used as biodegradable plastic bags. Moreover, Ghasemlou *et al.* [2013] reported the thickness of the edible film of corn starch was around 0.15 mm while Fakhouri *et al.* [2013] obtained 0.053 to 0.063 mm for those made from potato, rice, wheat, gelatin, and sorghum starch.

### Water and oil holding capacity

The WHC and OHC of NAS and PAAS bioplastics are shown in Figure 2. The WHC and OHC of NAS bioplastic were significantly different ( $p \leq 0.05$ ) from those of the PAAS ones. The highest WHC was found for NAS bioplastic. The PAAS bioplastics had lower WHC, which probably was associated with the presence of cross-linking bonds in the PAAS molecules hindering water penetration. However, the concentration of the STMP/STPP mixture used to obtain

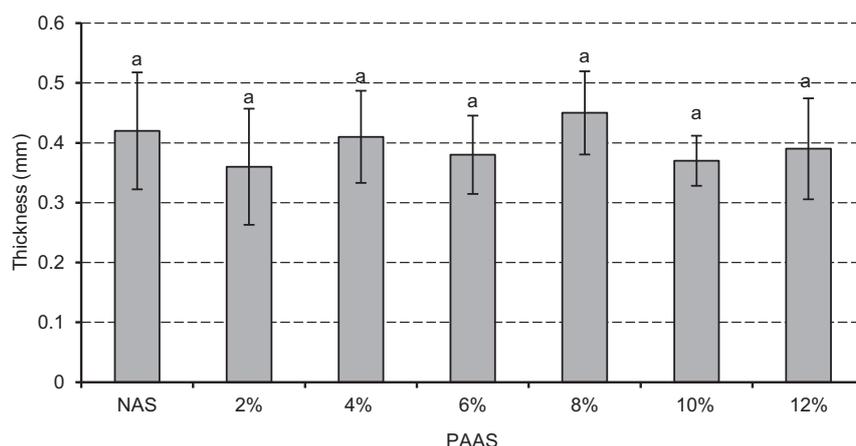


FIGURE 1. Thickness of bioplastics of native arenga starch (NAS) and phosphate acetylated arenga starches (PAAS) obtained using different concentrations of sodium trimetaphosphate/sodium tripolyphosphate (STMP/STPP) mixture (2–12%).

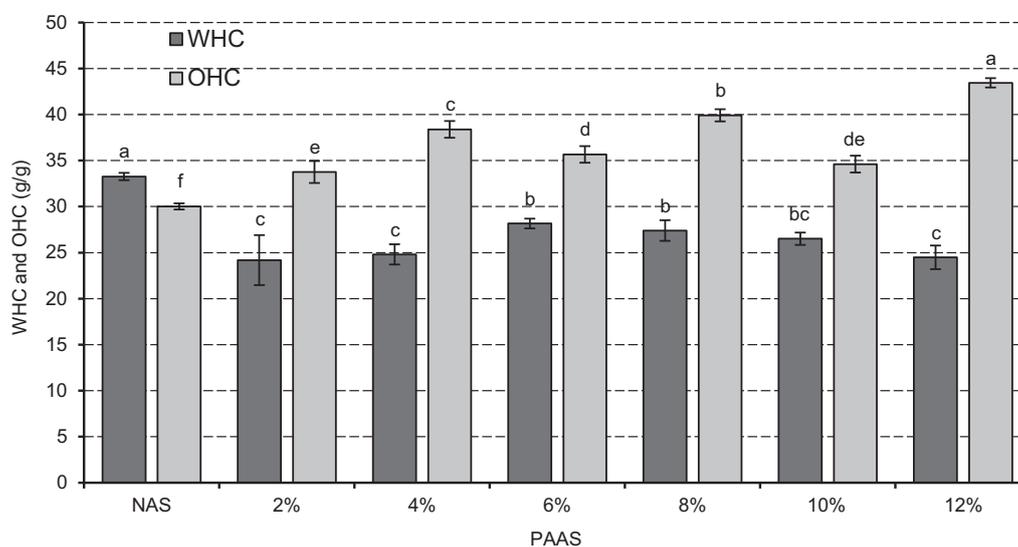


FIGURE 2. The water holding capacity (WHC) and oil holding capacity (OHC) of bioplastics of native arenga starch (NAS) and phosphate acetylated arenga starches (PAAS) obtained using different concentrations of sodium trimetaphosphate/sodium tripolyphosphate (STMP/STPP) mixture (2–12%). Different letters a-f above bars indicate significant differences among values ( $p \leq 0.05$ ).

the modified polymers only slightly differentiated the WHC of PAAS bioplastics. Meanwhile, the highest and lowest OHC were found for 12% PAAS and NAS bioplastics, respectively. Furthermore, the OHC tended to be increasing with the increased concentrations of the STMP/STPP mixture.

The WHC results are in agreement with the study that showed the water uptake of bioplastics from cassava peel starch reinforced with microcrystalline cellulose (Avicel PH101 fillers) was lower compared to the original material. This was attributed to the strong hydrogen bonds between microcrystalline cellulose and the molecular structure of starch [Maulida *et al.*, 2016]. The OHC results were in accordance with an earlier report of Sondari & Iltizam [2018] who showed that the OHC of bioplastics from modified cassava starch was lower than that of the original material.

#### Water vapor transmission rate

WVTR of the NAS and PAAS bioplastics is presented in Figure 3. The highest WVTR ( $2.40 \text{ g/h/m}^2$ ) was noted for

the bioplastic obtained using NAS. The lowest value ( $1.05 \text{ g/h/m}^2$ ) was determined for 4% PAAS sample, however this value did not differ significantly ( $p > 0.05$ ) from those determined for 2% and 6% PAAS bioplastics. It is important to note that the values obtained for the PAAS bioplastics were lower than those determined for NAS and this could be associated with the rigidity of PAAS which has the ability to prevent water from entering the starch molecules. Moreover, the incorporation of acetyl groups into the starch molecules has led to a reduction of OH groups and thus to the lower ability to bind water [López *et al.*, 2011]. This is in accordance with an earlier report of Detduangchan *et al.* [2014] that the WVTR of the modified rice starch bioplastic was lower than that of the native material.

According to Fakhoury *et al.* [2012], the water vapor permeability of the native cassava starch film was higher with  $4.88 \text{ g mm/m}^2 \text{ d kPa}$  compared to acetylated and crosslinked cassava starch, which was found to be  $3.59 \text{ g mm/m}^2 \text{ d kPa}$ . This phenomenon was caused by the strong interaction be-

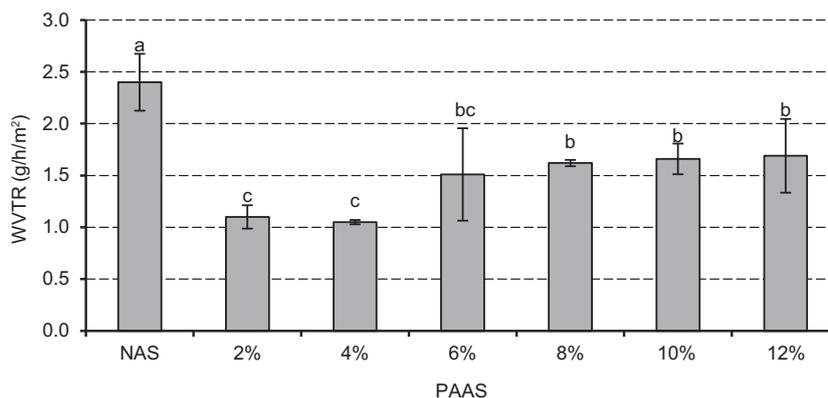


FIGURE 3. The water vapor transmission rate (WVTR) of bioplastics of native arenga starch (NAS) and phosphate acetylated arenga starches (PAAS) obtained using different concentrations of sodium trimetaphosphate/sodium tripolyphosphate (STMP/STPP) mixture (2–12%). Different letters a-c above bars indicate significant differences among values ( $p \leq 0.05$ ).

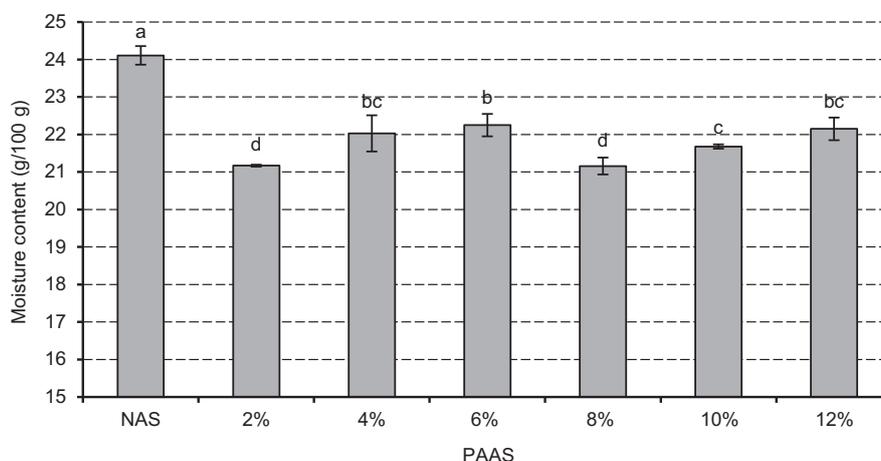


FIGURE 4. Water content of bioplastics of native arenga starch (NAS) and phosphate acetylated arenga starches (PAAS) obtained using different concentrations of sodium trimetaphosphate/sodium tripolyphosphate (STMP/STPP) mixture (2–12%). Different letters a-d above bars indicate significant differences among values ( $p \leq 0.05$ ).

tween amylose and amylopectin in the modified starch molecules. However, bioplastics with a low WVTR value are suitable for packaging food products to avoid damage caused by the surrounding environment.

#### Water content

The water content of the NAS and PAAS bioplastics obtained using different concentrations of the STMP/STPP mixture is shown in Figure 4. The concentration of STMP/STPP mixture significantly ( $p \leq 0.05$ ) affected the water contents in PAAS bioplastics. The highest value (24.11 g/100 g) was obtained for NAS bioplastic, while the lowest (21.16–21.17 g/100 g) was found for 2% and 8% PAAS bioplastics. Moreover, the water content in PAAS bioplastics was found to be generally lower than in NAS. It was supported by the findings of Gutiérrez *et al.* [2015] that the water content in phosphated corn starch edible film was 30%, compared to 44% determined in the native corn starch. However, Atef *et al.* [2015] reported that bioplastics were expected to have a low water content in order to ensure they do not increase the amount of water in the product when used for food packaging.

#### Biodegradation

The biodegradability of the bioplastic materials was tested using the soil burial method. Biodegradation is defined as a process of decomposition through the activities of microbes in the soil which leads to the transformation of a compound's structure thereby causing changes in molecular integrity [Liu *et al.*, 2016]. The average values of weight loss for the NAS and PAAS bioplastics are presented in Figure 5. The differences between the values were significant ( $p \leq 0.05$ ).

The weight loss was the highest in NAS bioplastic and lower in PAAS samples. However, for PAAS bioplastics, the values increased with the increasing concentrations of the mixture of STMP/STPP used to obtain the modified polymers. This shows NAS bioplastics are more easily degraded compared to PAAS bioplastics. A previous study showed 71% of potato peel bioplastic degraded in moist soil within four weeks [Arikan & Bilgen, 2019]. Another research reported the biodegradation of bioplastics made from native corn and rice starches and those modified was influenced by temperature, humidity, and microbial activity such that 48.73% was achieved after the materials were placed in the soil at a depth of 3 cm for 15 days [Marchelvam *et al.*, 2019].

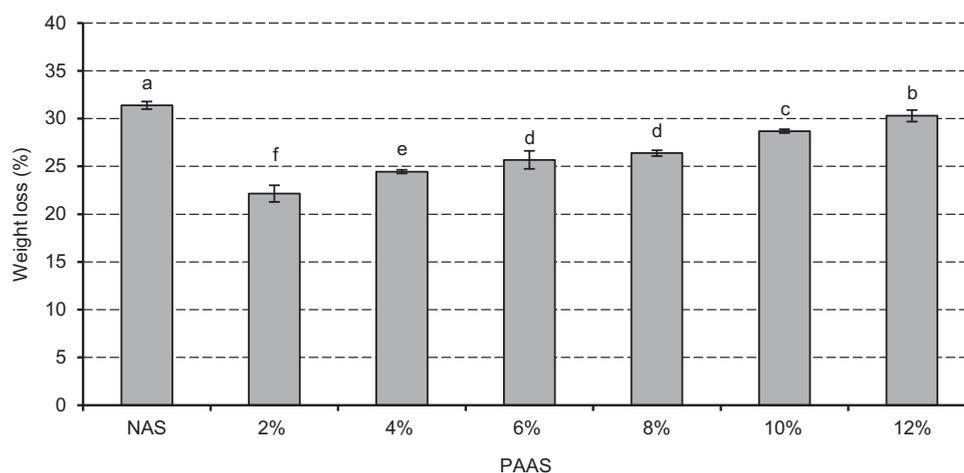


FIGURE 5. Weight loss of bioplastics of native arenga starch (NAS) and phosphate acetylated arenga starches (PAAS) obtained using different concentrations of sodium trimetaphosphate/sodium tripolyphosphate (STMP/STPP) mixture (2–12%). Different letters a-f above bars indicate significant differences among values ( $p \leq 0.05$ ).

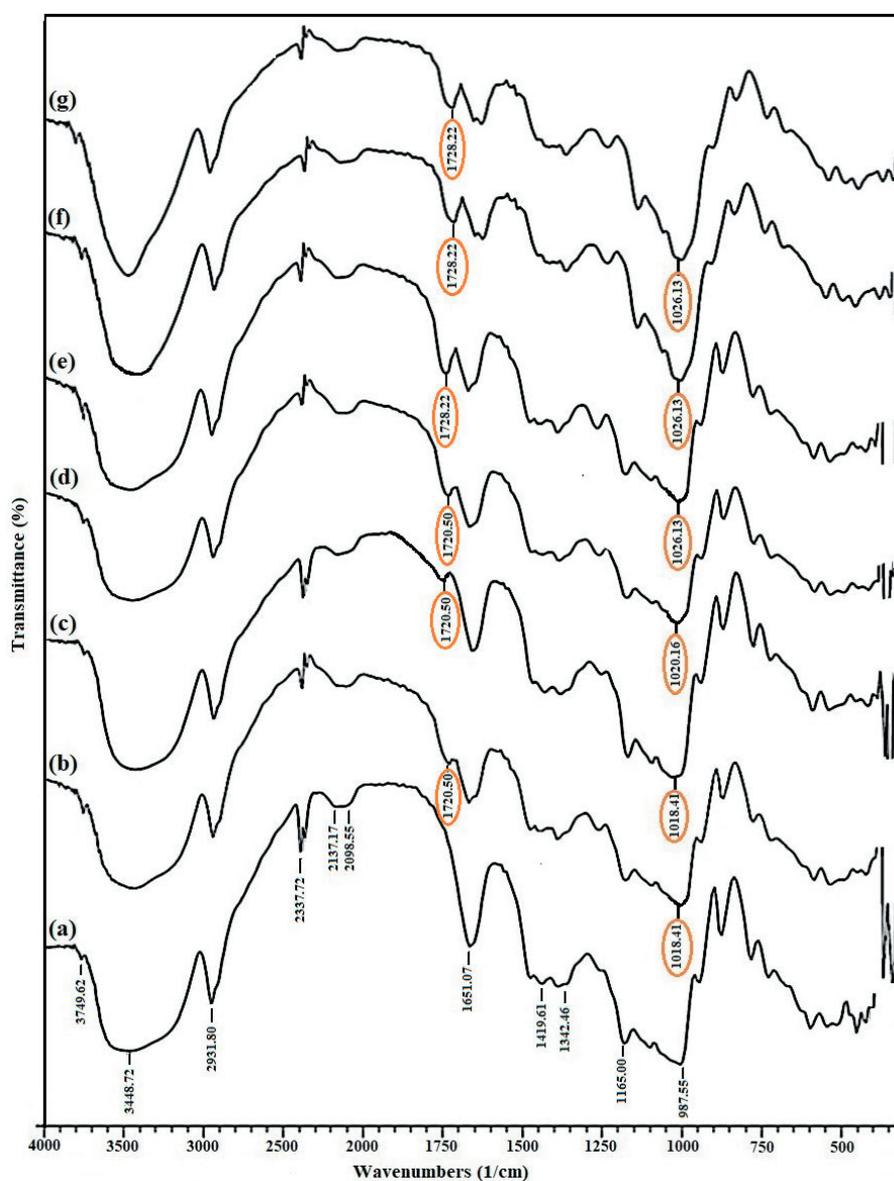


FIGURE 6. The FTIR spectra of bioplastics of native arenga starch (a) and phosphate acetylated arenga starches obtained using different concentrations of sodium trimetaphosphate/sodium tripolyphosphate (STMP/STPP) mixture: 2% (b), 4% (c), 6% (d), 8% (e), 10% (f), and 12% (g).

### FTIR spectra

FTIR spectra of NAS and PAAS bioplastics are shown in Figure 6. The characteristic bands were observed at the range of  $3700\text{ cm}^{-1}$  to  $3000\text{ cm}^{-1}$ ,  $2931.80\text{ cm}^{-1}$  and  $1651.07\text{ cm}^{-1}$  for the hydroxyl groups (O–H), methylene (C–H), and residual bound water ( $\text{H}_2\text{O}$ ) stretching the vibration of the glucose unit, respectively. In comparison with the values obtained for NAS bioplastic (Figure 6a), the new absorption bands in the range of  $1728.22\text{ cm}^{-1}$  to  $1720.50\text{ cm}^{-1}$  and  $1026.13\text{ cm}^{-1}$  to  $1018.41\text{ cm}^{-1}$  appeared for PAAS bioplastics (Figures 6b–g). These bands corresponded to C=O stretching vibration of an ester group and P–O–C stretching vibration, respectively. This confirmed the occurrence of acetylation and crosslinking between the starch molecules. Previously, acetylation of arenga starch was shown by the presence of the ester carbonyl group bands at  $1720\text{ cm}^{-1}$  [Rahim *et al.*, 2017]. In turn, Detduangchan *et al.* [2014] reported a peak at  $1035.75\text{ cm}^{-1}$  corresponding to the phosphate stretching (P–O–C) in STMP/STPP cross-linked rice starch films.

### Mechanical properties

The tensile strength, elongation at break, and Young's modulus were determined to evaluate the mechanical properties of NAS and PAAS bioplastics. Tensile strength is the magnitude of the force required to achieve the maximum pull in each bioplastic area. Elongation at break is the percentage change in bioplastic length calculated when it was pulled up to break, whereas the Young's modulus represents the division of the tensile strength by the elongation at break [López *et al.*, 2016]. The average values of these parameters for NAS and PAAS bioplastics are shown in Table 1. The ANOVA indicated that tensile strength of the samples did not differ significantly ( $p > 0.05$ ), while analysis differentiated bioplastics in terms of elongation at break and Young's modulus ( $p \leq 0.05$ ).

Generally, the elongation at break of the PAAS bioplastics was higher than that of NAS bioplastic, although it should be noted that the values determined for NAS and 2%, 4%, 12% PAAS did not differ significantly ( $p > 0.05$ ). Zhang *et al.*

[2013] reported an increase in elongation at break of oxidized corn starch films due to an increase in the mobility of starch chains resulting from a stronger interaction between oxidized starch and starch chains and a higher degree of oxidation. Moreover, Woggum *et al.* [2014] also found the elongation at break of dual modified rice starch with a mixture of STMP 2% – STPP 5% was higher than that of the native starch.

Furthermore, the elongation at break of the PAAS bioplastics increased with the concentrations of STMP/STPP up to 6% and subsequently remained constant at 12%. Meanwhile, the Young's modulus of the PAAS bioplastics decreased with the concentrations of STMP/STPP up to 8% and afterwards remained constant at 12%. However, the elongation at break and Young's modulus of bioplastics from chitosan cross-linking methylcellulose using STMP 0.1 and 0.3% were found to be higher than these of the chitosan bioplastics [Wang *et al.*, 2019].

### Sensory attributes

The degree of likeness of color, texture, aroma, and overall acceptability of the NAS and PAAS bioplastics were evaluated and the results are presented in Table 2. The color and texture were significantly different ( $p \leq 0.05$ ), while the differentiations among bioplastics in aroma and overall acceptability were not significant ( $p > 0.05$ ).

The color and texture of the PAAS bioplastics were more transparent and their surface was smoother compared to NAS bioplastic, respectively. The transparency and smoother surface of the PAAS bioplastics were achieved through acetylation and crosslinking. In line with the findings of this research, it was reported that the color of the bioplastics from oxidized, crosslinked, and dual oxidation crosslinked lotus rhizome starch was clearer compared to the native material [Sukhija *et al.*, 2019]. This is in agreement with the report by Wang *et al.* [2019] who showed the colors and texture of films made from chitosan methylcellulose crosslinking were more transparent and smoother due to the compactness of the starch molecules.

TABLE 1. The tensile strength, elongation at break, and Young's modulus of the NAS and PAAS bioplastics obtained using different concentrations of STMP/STPP mixture.

| Bioplastic | Tensile strength (MPa) | Elongation at break (%) | Young's modulus (MPa) |
|------------|------------------------|-------------------------|-----------------------|
| NAS        | $1.08 \pm 0.09^a$      | $7.03 \pm 0.83^b$       | $0.15 \pm 0.01^a$     |
| PAAS 2%    | $1.10 \pm 0.12^a$      | $7.87 \pm 1.28^{ab}$    | $0.14 \pm 0.01^{ab}$  |
| PAAS 4%    | $1.09 \pm 0.29^a$      | $7.78 \pm 1.45^{ab}$    | $0.14 \pm 0.04^{ab}$  |
| PAAS 6%    | $1.16 \pm 0.16^a$      | $9.14 \pm 1.09^a$       | $0.13 \pm 0.02^{ab}$  |
| PAAS 8%    | $0.94 \pm 0.07^a$      | $8.09 \pm 1.05^a$       | $0.12 \pm 0.01^b$     |
| PAAS 10%   | $0.93 \pm 0.03^a$      | $8.17 \pm 0.54^a$       | $0.11 \pm 0.01^b$     |
| PAAS 12%   | $0.99 \pm 0.11^a$      | $7.87 \pm 1.17^{ab}$    | $0.13 \pm 0.01^{ab}$  |

Data are mean  $\pm$  standard deviation (SD). Values in the same column with different superscript indicate a significant difference ( $p \leq 0.05$ ). NAS = native arenga starch, PAAS = phosphate acetylated arenga starches, STMP = sodium trimetaphosphate, STPP = sodium tripolyphosphate.

TABLE 2. Sensory assessment of the NAS and PAAS bioplastics obtained using different concentrations of STMP/STPP mixture.

| Bioplastic | Color                | Texture              | Aroma             | Overall acceptability |
|------------|----------------------|----------------------|-------------------|-----------------------|
| NAS        | $4.60 \pm 0.74^b$    | $4.60 \pm 1.12^b$    | $4.48 \pm 1.13^a$ | $5.33 \pm 0.72^a$     |
| PAAS 2%    | $5.40 \pm 0.63^a$    | $5.47 \pm 0.99^a$    | $4.60 \pm 0.83^a$ | $5.33 \pm 0.72^a$     |
| PAAS 4%    | $5.27 \pm 0.88^{ab}$ | $5.53 \pm 0.92^a$    | $4.73 \pm 0.70^a$ | $5.53 \pm 0.83^a$     |
| PAAS 6%    | $4.93 \pm 0.88^{ab}$ | $5.47 \pm 0.99^a$    | $4.53 \pm 0.64^a$ | $5.60 \pm 0.63^a$     |
| PAAS 8%    | $5.67 \pm 1.11^a$    | $5.27 \pm 0.96^{ab}$ | $5.07 \pm 0.88^a$ | $5.47 \pm 0.64^a$     |
| PAAS 10%   | $5.13 \pm 0.99^{ab}$ | $5.20 \pm 0.86^{ab}$ | $5.07 \pm 0.96^a$ | $5.20 \pm 0.68^a$     |
| PAAS 12%   | $5.07 \pm 1.03^{ab}$ | $5.13 \pm 0.74^{ab}$ | $4.73 \pm 1.03^a$ | $5.20 \pm 0.68^a$     |

Data are mean  $\pm$  standard deviation (SD). Values in the same column with different superscript indicate a significant difference ( $p \leq 0.05$ ). NAS = native arenga starch, PAAS = phosphate acetylated arenga starches, STMP = sodium trimetaphosphate, STPP = sodium tripolyphosphate.

## CONCLUSION

The dual modification of NAS by acetylation and phosphorylation using STMP/STPP mixture with 2–12% concentrations proved to be an advantageous strategy to obtain the PAAS bioplastics with improved physicochemical, mechanical, and sensory properties compared to NAS bioplastic. The WHC, WVTR, moisture content, and Young's modulus of the PAAS bioplastics were generally lower than these of the NAS bioplastic, while the OHC and elongation at break of the PAAS bioplastics were higher compared to NAS bioplastics. The NAS and PAAS bioplastics were decomposed in the soil. Furthermore, the color and texture of the PAAS bioplastics were more transparent and smoother than these of the NAS bioplastic, respectively. This has indicated that the PAAS bioplastics were superior over the NAS bioplastic.

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## CONFLICT OF INTERESTS

The authors declare there is no conflict of interest.

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