PJN
ISSN 1680-5194

PAKISTAN JOURNAL OF NUTRITION

ANSInet
308 Lasani Town, Sargodha Road, Faisalabad - Pakistan
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OPEN ACCESS Pakistan Journal of Nutrition

ISSN 1680-5194
DOI: 10.3923/pjn.2019.42.50

Research Article

Physicochemical Properties of Modified Palado Seed Flour (*Aglaia* sp.) from Pregelatinization, Cross-Linking and Acetylation

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Abstract

Background and Objective: The palado seed flour increase the economic value and shelf life of processed palado products. The aim of this study was to know the physicochemical characteristics of modified palado seed flour. **Materials and Methods:** This study was conducted in three stages: The first stage was flour production from palado seed, the second stage was flour modification using pregelatinization, cross-linking and acetylation and third the stage was analysis of physical and chemical properties. These analyses included proximate content analysis, such as evaluation of the carbohydrate, energy, starch, amylose and amylopectin contents. Meanwhile, the physical properties evaluated include the yield, degree of whiteness, density, heat viscosity and cold viscosity. **Results:** The results of the analysis of the physicochemical properties of modified palado seed flour showed a water content (6.72-8.23%), an ash content (1.25-3.26%), a fat content (9.46-9.75%), a protein content (5.46-8.75%), a carbohydrate content (71,04-73.50%), an energy content (402-417 kcal), a starch content (57.99-63.98%), an amylose content (0.44-0.85) and an amylopectin (57.44-63.13%). The resulting yield was in the range of 60.35-68.91%, the degree of whiteness was 45.23-61.69%, of the density was 0.59-0.69 g mL^{-1}, the heat viscosity was 6.20-524.00 cP and cold viscosity was 6.45-313.00 cP. **Conclusion:** It is concluded that palado seed flour is useful as an alternative flour based on its physicochemical properties, before and after modification.

Key words: Palado seed flour, flour technology, food sovereignty programs, starch, alternative flour

Received: April 17, 2018 Accepted: September 09, 2018 Published: December 15, 2018

Citation: Syamsul Rahman, S. Salengke, Abu Bakar Tawali and Meta Mahendradatta, 2019. Physicochemical properties of modified palado seed flour (*Aglaia* sp.) from pregelatinization, cross-linking and acetylation. Pak. J. Nutr., 18: 42-50.

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Competing Interest: The author has declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

In supporting food sovereignty programs, flour technology is the best solution. The study aimed to develop a new material source (beside wheat) for use as breads material; noodles and other food products. Since 1997, the government's subsidy towards the flour had been eliminated; therefore, the price has increased with flour consumption[1]. The needs for flour are 20% for bread, 50% for noodles (instant noodles, dry noodles and wet noodles inside the small industry), 10% for biscuits and snack and the rest for household use[1]. Thus, a diversification effort with local flour needs to be developed.

Food diversification needs to be implemented to support government food sovereignty programs[2]. One of the ways to implement food diversification is to dig out, utilize and socialize the potential of plasma nutfah, which has not been before. Indonesia is well-known for its wealth of biological diversity, which is able to satisfy the needs of human. The utilization of plants as an alternative food source are needed to reduce dependence on the main food, particularly rice, as the source of carbohydrate[2]. The development of carbohydrate-based foods is still dominated by 70% of flour coming from rice, 15% of flour coming from tubers and local seeds and 15% of flour coming from wheat[3].

One plant material containing a high carbohydrate content is palado seed (*Aglaia* sp). In addition to its high carbohydrate content, this plant is one of the wild plants that grows in moist forests to ages, of hundreds of years and heights of approximately 30-50 m. On average, trees over 10 years old can produce 250-500 kg of wet palado fruits. The seed of palado fruit contains 64.04-64.91% carbohydrates, thus, it has a potential as an alternative food source[4]. In addition to carbohydrates, palado seeds also contain fat (14.75-14.88%), protein (8.94%-8.95%), water (4.87-5.59%), ash (3.66-3.71%) and crude fiber (3.74-3.77%)[4]. Picture of the trees, fruits and seeds of palado are shown in Fig. 1.

Aglaia sp is member of the *Meliaceae* family. This plant is mostly found in Sulawesi, especially western Sulawesi. The distribution area in western Sulawesi is quite wide, including river flow areas; such as the Karama River, Karataun River and Bonehau River; valley areas and Mamuju Regency Mountains, including North Mamuju, Majene, Polman and Mamasa. *Malanesia* regions have numerous *Aglaia* types, such as Borneo (50 kinds), while 38 types are found in Sumatera[5]. Regarding the utilization of certain types of *Aglaia* sp its wood is utilized as construction material. The fruit can be eaten, while the flower of *A. Odorata* is used as tea and perfume material due to their aromatic smell[5].

Thus, the palado seed flesh has potential as a food source. However, to date, research on the physicochemical properties of modified palado flour has never been performed. Therefore, the palado flour needs to be developed through a modified method, including one pregelatinization, cross-linking and acetylation. This study aimed to know the physicochemical characteristics of modified palado seed flour.

Fig. 1(a-d): (a) Small palado plants, (b) Producing palado plants, (c) Palado fruit and (d) Palado seeds

MATERIALS AND METHODS

Materials and tools: The main ingredient used is palado fruit obtained from the forests around the river flow areas of Karama, Sampaga District, Mamuju Regency and West Sulawesi Province. Level of ripeness of these fruits was optimal, because they fell from the trees. The chemical materials used for analysis were obtained from the Laboratory of Food Science and Technology of the Agriculture, Faculty in Hasanuddin University (UNHAS) and the Laboratory of Food Science and Technology, Department of the Faculty of Agriculture Technology, Institute Pertanian Bogor (IPB). The study was divided into four steps. The first step was making flour from palado seeds. The second step was starch modification using pregelatinization, cross-linking and acetylation. The third step was proximate content analysis and analysis of the chemical properties of the flour. The fourth step was analysis of the physical properties of palado seed flour.

Making palado seed flour: The flour was made by adopting a method previously performed[6] to make corn flour. This process started by soaking dried palado seeds in water for approximately 12 h. Then, the palado seeds were sieved and milled using a disc mill. The flour was then heated in a cabinet dryer for 15-18 h at 50-60°C until dry. Next, the flour was sieved using a 60 mesh sifter, with a 0.250 mm particle size. The sucrose content in the resulting flour was determined as previously described[7] to be 84.21%. The samples of palado seed flour totaled 800 g, consisting of 200 g of flour without modification, 200 g for pregelatinization; 200 g for cross-linking and 200 g for acetylation.

Making modified palado flour: Palado seed flour was modified in three steps, pregelatinization, cross-linking and acetylation. Pregelatinization[8], cross-linking[9] and acetylation[10], were performed as previously described.

Proximate content analysis and chemical properties of the flour: Chemical properties of the palado seed flour consists of proximate content analysis that includes analysis of the water level using an oven as described by AOAC[11] and modified by Kuswandari *et al.*[12]. The level of ash was analyzed by burning in an oven[11]. The fat and levels were determined using the Kjeldahl micro-method[11]. The starch level was analyzed using the Trimetric method described by the Luff-Schoorl technique[13]. The amylase was analyzed spectrophotometrically[14]. The carbohydrates level was determined using the difference method[15] and the energy was calculated by multiplying the total weights (grams) of crude protein, carbohydrate, total fat and food fiber by factors of 4, 4, 9 and 2 respectively[16].

The physical properties of palado seed included the sucrose content, which was determined as previously described by Akanbi *et al.*[7]. The degree of whiteness was determined using a whiteness meter[17]. The hot viscosity and cold viscosity were determined using a viscometer[18]. The density was determined using a gravimetric method[12].

Data analysis: Data from the analysis of the physicochemical properties of palado seed flour obtained in the 3 treatments were averaged. To determine each treatment parameter, the standard deviation was calculated. The data were presented in a table form and; then analyzed using descriptive analysis via; both quantitative and qualitative approaches.

RESULTS AND DISCUSSION

Analysis of the proximate composition and chemical properties of the flour: The proximate composition and chemical properties of modified palado flour that were analyzed include the water, fat, ash, energy, carbohydrate, starch and amylose contents and the result are shown in Table 1.

Table 1: Proximate composition and chemical properties of modified palado seed flour (100 g)

	Modified flour			
Characteristics	Unmodified	Pregelatinized	Cross-linking	Acetylation
Water content (%)	8.35±0.68	6.87±0.71	8.23±0.70	6.72±0.71
Ash content (%)	3.61±0.69	3.26±0.69	3.27±0.71	1.25±0.69
Crude fat (%)	12.37±0.71	9.46±0.70	9.54±0.71	9.75±0.68
Crude protein (%)	8.91±0.70	9.37±0.71	5.46±0.69	8.97±0.70
Carbohydrat (%)	66.72±0.70	71.04±0.70	73.50±0.70	73.31±0.70
Energy (kcal)	414.00±0.69	407.00±0.69	402.00±0.69	417.00±0.71
Starch (%)	52.78±0.71	63.98±0.69	59.40±0.70	57.99±0.71
Amylose (%)	1.00±0.68	0.85±0.71	0.44±0.71	0.53±0.69
Amylopectin (%)	51.78±0.70	63.13±0.70	58.96±0.70	57.44±0.70

The number after the sign (±) is the standard deviation value

Water content of palado flour: From the research results, it was found that the water content of unmodified flour was 8.35% while that of the modified samples was approximately 6.72-8.23%. This showed that the water content in modified palado flour was still lower than that in unmodified palado flour and the water content in wheat flour was previously reported[19], to be 11.32%. However, water content in modified palado flour was still higher than that previously reported in sweet potato[16].

The highest water content in modified palado flour (8.23%) was found upon cross-linking. The high water content upon cross-linking was caused by the phosphate group that were adhered to starch granules and these groups were ionic and thus able to bind the water[20]. Moreover, the lowest water content in modified palado flour (6.72%) was with acetylation modification.

This finding supports the opinion of Erika[21] who stated that modifying starch using acid hydrolysis (acetylation) results in starch with a looser structure so water can be easily evaporated during drying. Meanwhile, the water content in pregelatinized flour was 6.87%. The decreasing water content in pregelatinized flour was caused by the parboiling pregelatinization treatment, which applied heat and caused water absorption as well as swelling of starch granules[22]. When the gelatinized flour had dried, the water could be easily removed from the hydroxyl bond so the water content decreased slightly.

The total water content in the ingredients will influence the resistance to damage caused by microbes or insects[23]. Flour is dried to decrease the water content to a certain limit to block the growth of microbes and the activity of enzymes, which cause damage to flour. The water content limit for which microbes can be grown is approximately 14-15%.

Ash content of palado flour: The ash content represents the content of inorganic materials inside food, especially modified flour. The ash content found in this study were 1.25-3.26%, while the level of ash content in unmodified palado flour was 3.61%. The lowest ash content in modified palado flour (1.25%) was with acetylation modification. The ash content is influenced by extraction and by washing the flour/starch using water, as the minerals dissolved in the water are easily discarded along with the dregs[24].

The highest level of ash found in palado flour (3.27%), was with cross-linking modification. This happened due to the increasing number of tied phosphate groups because phosphate is a component of the ash structure[20]. This finding is consistent with previous findings[25] that phosphorization could increase the ash content in pulverized rice.

This is caused by the decrease in the starch level and by the addition of phosphorous (P) into the flour. The increasing P content in this study was used as an indicator of the success of the phosphorization process, considering that P is the bridge in cross-linking that is formed in this process[25]. The ash content of palado flour was higher than the ash content in wheat flour (0.84%) and jarring seed flour (1.34%)[19].

Quantitatively, the ash content in starch comes from the minerals inside the seeds as well as from ground and air contamination during production. The production of food can also influence the availability of minerals inside the food[26]. The use of water during washing and soaking can decrease the availability of minerals because minerals are able to be dissolved in the water used. The extraction process, when used on food can truly decrease the mineral content.

Fat content of palado flour: The results show that differences in the modification method did not lead to any real differences in the fat content, which was approximately 9.46-9.75%. The fat content of flour without modification (natural flour) was 12.37%. Indirect modification can influence the fat content but influences the starch content more[24]. Moreover, changes in the starch structure have also been reported, which occurs due to modification of cassava because starch does not hydrolize amylose; this structure is suspected to maintain the fat inside the amylose polymer chain such that it does not result in fat level differences among the modification treatments[24].

Generally, modified palado flour contains more fat than does wheat flour, which contains 1.57%[19] and fat contents of 1.00 and 1.00% have been found in pulverized rice and tapioca flours respectively[27]. Alternatively[28] the fat content in corn starch was reported to be 0.6-0.8%, in wheat starch was 0.8-1.2% and in rice starch was 0.6-1.4%. However, a high fat content can be considered a nutritional factor, that is considered less advantageous for starch storage process because fat can cause rancidity[29].

According to previous report[23], the fat content inside starch can disturb the gelatinization process because fat is able to form a complex using amylose so it can block the amylose from leaving the starch granule. In addition, most fat can be absorbed by the granule surface and thus, will form a hydrophobic fat layer around the granule. This fat layer will block the water bonded by the starch granule. This affects the viscosity and the stickiness of starch will decrease due to the decrease in the water content.

Protein content of palado flour: The protein content of modified palado seed flour was 5.46-8.97%, while that of unmodified palado flour was 8.91%. According to a previous report[24], an increase in the temperature can cause a decrease in the starch level so that the starch protein content will relatively increase. The protein content of modified palado flour was still higher than that of sweet potato flour, which was reported in the range of 3.50-3.85%[19].

It is not expected that flour or starch with a low protein content will experience a decrease in flour viscosity so that flour quality will decrease[23]. Protein and starch can form a complex on the granule surface and cause a decrease in the viscosity, which will cause a low strength of the gel. In content starch, the protein content in starch is expected to be higher. This is related to the use of starch; when the protein content is higher, no other substitute ematerial is required.

Contents of carbohydrates and energy in palado flour: The results of this study show that the modified palado flour had 71.04-73.50% carbohydrates. and 402-417 kcal of energy. Alternatively, the carbohydrate and energy contents of unmodified palado flour were 66.72% and 414 kcal, respectively. As observed regarding the energy content, the energy of palado flour was higher than that of wheat flour (365 kcal), pulverized rice (364 kcal) and corn flour (355 kcal)[30]. The energy content in sweet potatoes, which was reported[16] to be between 392.00 and -399.60 kcal was similar. However, the carbohydrates content was lower than that in sweet potato flour (77.3%), pulverized rice (80.0%) and corn flour (73.30%)[30], while that in sweet potato flour was 86.35-88.40%[16].

Starch content in palado flour: Starch content is one of the quality criteria for flour, either as a food or nonfood material[23]. This study obtained a starch content of 52.78% in unmodified palado flour, while modified palado flour had a starch content of 57.99-63.98%. The highest starch content was obtained through pregelatinization and was approximately 63.98% and the lowest value (57.99%) was with acetylation. This result agrees with Wulan *et al.*[20], who stated that the starch content upon discontinued combined branch bonding with cross-linking was higher than that in natural starch. This finding was observed because the phosphate groups penetrated into the granule, forming covalent bonds with the starch molecule, which resulted in a larger molecule and thus, highlighted the whole starch molecule.

The starch content in modified palado flour was higher than that in gadung starch (*Dioscorea hispida*), which was 38.80%[31] and the content of canna was approximately 40.18%[23]. Anggraeni and Yuwono[32] also reported the results of the analysis of starch in the main sweet potato material (*Ipomoea batatas)* and found that the content of starch in the yellowish red variety was 23.55%, that in the yellowish white variety was 22.78% and that in the *Ayamurasaki* (purple) variety was 16.37%.

The starch content in the main ingredient can be influenced by the age of plants and the duration of storage after harvest[32]. The factors that influence the drying process of food are the initial water content of the ingredient, the humidity of the environment and the mediator of heat movement[31].

Amylose and amylopectin content in palado flour: The contents of amylose and amylopectin in unmodified palado flour obtained in this study were 1.00 and 51.78%, respectively, while those in modified palado flour was approximately 0.44-0.85 and 57.44-63.13% respectively. This showed that the ratio of amylose to amylopectin in modified palado flour was 1:5. According to a previous study conducted by Munarso *et al.*[25], the amylopectin molecule can more easily undergo phosphorization (cross-linking) than can the amylose molecule. Therefore, the amylopectin molecules join each other, resulting in fewer molecules with a large size. This causes the proportion of amylose in comparison amylopectin to be increased. Additionally, in a previous study, Hapsari *et al.*[22] also stated that pregelatinization slightly decreased the content of amylose in cassava flour. This finding was observed because the starch was heated in water at the gelatinization temperature and the heat energy caused the hydrogen bonds in the starch to weaken. The weaker bond will ease the water entering the starch and generate little dissolution and the amylose molecule changed in the water.

There are differences between types of starch producers, such as differences in amylose and amylopectin, including the structure and content, granular organization, presence of lipids, proteins and minerals and size of the starch granules[28]. This also explains the ratio of amylose and amylopectin in starch granules, which is truly important and is often the determining parameter in selecting the starch source to be applied in the production process of food to obtain desired functional properties[33]. This finding was because the ratio of amylose and amylopectin could influence the ability of starch to form a gel, thicken and form a film.

The components of amylose are related to the increasing water absorption ability and the perfection of the gelatinization process, while the amylopectin content determines the ability to develop the product[34]. Moreover, the contents of amylose and amylopectin in starch can influence the size of the starch granules and weight of the starch molecules. At the same time, amylopectin may play a role in

Table 2: Physical properties of modified palado seed flour (100 g)

	Modified flour			
Characteristics	Unmodified	Pregelatinization	Cross-linking	Acetylation
Yield (%)	84.21±0.71	65.26±0.69	68.91±0.70	60.35±0.69
Whiteness (%)	29.27±0.70	45.23±0.71	61.69±0.69	52.59±0.70
Density (g mL^{-1})	0.49±0.70	0.67±0.70	0.69±0.71	0.59±0.71
Viscosity 85°C (cP)	2.64±0.69	6.20±0.70	524.00±0.70	28.85±0.70
Viscosity 30°C (cP)	2.73±0.70	6.45±0.69	313.00±0.71	12.90±0.71

Note: The number after the sign (±) is the standard deviation value

the puffing process[35]. The food coming from starch with a high amylopectin is light, crunchy and crispy. Generally, the starch of food contains 25% amylose and 75% amylopectin[35].

Analysis of the physical properties of palado flour: The physical properties of palado flour that were measured included the yield, whiteness, density, hot viscosity and cold viscosity and the result are shown in Table 2.

Yield: The results showed that the yield of unmodified palado flour (control) was 84.21%, while the yields of modified palado flour was 60.35-68.91%. The highest sucrose content in modified palado flour compared to the control palado flour was upon cross-linking. According to a previous study conducted by Wulan *et al.*[20], cross-linking causes bonds between amylose and amylopectin chains and between amylopectin chains. The greater the cross-linking is, the stronger the structure of the starch molecule. Afterwards, cross-linking can stabilize and strengthen the structure of the starch granule[20]. Therefore, a decrease in the amount of starch can be blocked, which will then result in a higher yield.

On the other hand, acetylation or extrusion of palado flour resulted in the lowest yield, which was 60.35%. According to a previous study, extrusion damages the starch[20]. Decreasing the amount of starch will automatically decrease the yield of the extruded starch[20].

Whiteness of palado flour: Whiteness is one of the physical parameters that is important to identify flour color[29]. The average whiteness values of modified palado flour were 45.23-61.69%, while the control unmodified palado flour had a value of approximately 29.72%. Compared with the other three treatments of palado flour modification, the lowest whiteness, (45.23%) was upon pregelatinization. According to Hapsari *et al.*[22], this result is caused by the heating process in pregelatinization, which will dissolve some chemical components inside the flour and the starch cell such as sugar, amylose, protein and preereduction sugar, which react and result in a brownish pigment that decreases the whiteness. These results agree with Darmajana[36], who explained that the main problem in processing dry food from high carbohydrate-containing food is the browning reaction. The browning reaction occurs during drying and causes the appearance of a brown color.

Alternatively, the palado flour samples with cross-linking and acetylation modification showed whiteness values of 61.69 and 52.59%, respectively. This result agrees with Nur and Purwiyanto[37], who found that using sodium hypochlorite in the chemical modification of starch, which will have a whitening effect on the resulting starch product because sodium hypochlorite is an oxidizer and takes on the role of as whitener. In addition, the acetylated oxidized starch has a higher whiteness because the acid during the acetylation process will increase the degree of whiteness and the use of sodium hypochlorite will increase the whiteness.

This study indicated that modified palado flour had a higher whiteness than *Dioscorea alata,* which has a value of 20.05% and *Amorphophallus campanulatus*, which had a value of 39.05%[23]. Nevertheless, the brightness of modified palado flour was lower than the quality standard of starch based on the Indonesian National Standard, which requires that the whiteness be at least 85%.

Palado flour density: Kamba density is a physical property of foods such as seeds and flours. According to a previous study, a material is considered kamba when the density is low[38]. Knowledge of the kamba density is needed, especially with regard to packaging, storage and transportation. The parameter of kamba density is mostly used to characterize foods, especially flours[39].

Unmodified palado flour had a kamba density value of 0.49 g mL^{-1}. This value was different from that of the modified palado flour, which had kamba density values of 0.59-0.69 g mL^{-1}. This shows that modified palado flour had a larger kamba density than the unmodified flour, with a kamba density of 0.49 g mL^{-1}. According to a previous study, a larger kamba density indicates that the weight unit of the same starch/flour needs a smaller storage space[38].

In a small movement space, the particles will not develop quickly because the kinetic energy of the particles is higher than that of the water molecules[39]. On the other hand, soaking will decrease the kamba density and increase the water level in the seeds[29]. Moreover, there are also reports that the heating of modified *Amorphophallus campanulatus* decreases the kamba density[12]. This finding is observed because the starch molecules start to be hydrated, which then weakens the interactions occurring on amylose and amylopectin; and thus will result in a small crystalline area and more domination by an amorphous area so that the granule structures will become irregular.

Hot viscosity and cold viscosity in palado flour: The cold and hot viscosities of modified palado flour in this study were measured using a Brookfield Viscometer. The hot viscosity was measured when the pasta was heated to 85°C and the cold viscosity was measured at a temperature of 30°C. The highest viscosity at 85°C was 524.00 cP. The viscosity was the maximum during the heating process. According to a previous study, the maximum viscosity is the point at which the starch granule expands and starts to break, followed by a decrease in the viscosity[39]. Moreover, heating causes the expansion of the granule, the cohesive capacity starts to weaken and the viscosity starts to increase[39].

In addition, the hot viscosity (95°C) is the criteria used to determine the starch granule capacity in defending itself or to determine the viscosity during the heating process[25]. Among the three modification methods of palado flour, the highest viscosity value was with cross-linking modification, for which the hot viscosity was 524.00 cP. This agrees with Munarso *et al.*[25], who stated that the phosphorization process increases the hot viscosity of pulverized rice. It also states that the hot viscosity of starch is influenced by many factors, such as the mixing effect of expanded starch granules, granule fragment, colloids formed from dispersed starch molecules, the development level of amylose and the competition to obtain free water among the floating amylose with the remaining granules[40].

Alternatively, the cold viscosity is the parameter used in the gel treatment of certain starches during cold conditions[25]. The highest viscosity at a temperature of 30°C was 313.00 cP, which occurred for cross-linked palado flour.

This agrees with a previous study conducted by Munarso *et al.*[25] who stated that the phosphorization process (cross-linking) is expected to result in starch with a colder viscosity. Therefore, the use of phosphorized starch is expected to prevent the syneresis process or the coming out of water from a gel matrix of a certain produced product. Furthermore, it has also been reported that an increase in viscosity occurs when the starch is hot (92.5°C) and faces the cooling process (40°C) which is caused by the tendency of starch to encounter retrogradation, where the characteristics are especially caused by hydroxyl groups[40]. The granule will expand due to the excellent gelatinization and will break and form thick mass[38].

CONCLUSION

It is concluded that palado seed flour is useful as an alternative flour based on its physicochemical properties, both before and after modification.

SIGNIFICANCE STATEMENT

This study presents new information regarding the existence of an alternative flour that has been found, which can be substituted for other flours. This study will help researchers uncover potential alternative sources of flour that have not been explored. This study is also an alternative solution for culinary entrepreneurs to obtain affordable flour based on local resources. In addition, the study can be used as a reference for researchers, students and culinary entrepreneurs.

ACKNOWLEDGMENTS

My thanks to: (1) Kemenristekdikti, for financial assistance provided in carrying out this research. (2) Chancellor of Makassar Islamic University (UIM) through LP2M UIM who has given direction and guidance and (3) Manager of ITP Laboratory, Faculty of Agriculture, UNHAS and ITP Laboratory Fateta IPB in conducting research.

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