

## EFFECT OF TEMPERATURE AND TIME ON DRY GRANULATION PROCESS OF ARENGA PALM SUGAR

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### ABSTRACT

The purpose of this study was to determine the physico-chemical, structure and morphology properties of arenga palm sugar blocks and granule and to analyse phenomenon on dry granulation of arenga palm sugar granule. Arenga palm sugar block was prepared by heating sap until temperature of  $118 \pm 2$  °C, then it was poured into wood mold and cooled to form solid state (arenga palm sugar block, GAC). The GAC, then was sized reduction and heated at temperature of 70, 80 and 90 °C and every 5 minutes it was stirred and observed its changes until 45<sup>th</sup> minute to produce arenga palm sugar granule (GAG). The arenga palm sugar granule by wet granulation was used as a control (GAG-control). Physico-chemical of arenga palm sugar block consisted of 10.1% db of moisture content, 3.2%db of reducing sugar and 92.5%db of sucrose was successfully used to produce palm sugar granule by dry granulation. The arenga palm sugar block with its crystallinity of 63-66% increased its crystallinity into 75% at 20<sup>th</sup> minute and then decreased into 72% at the end of the granulation process. Generally, morphological structure of arenga palm sugar block consisted of crystal particles wrapped by a dough-like binder. During granulation, the binder volume decreased due to moisture loss then granule surface was rougher than the initial one. Arenga palm granule by dry granulation had a higher transmittance than wet granulation, and dry granulation did not cause a dramatical change on functional group of the product. The granulation process (particle formation) occurs when moisture of the material reached around a multilayer water condition.

**KEYWORDS:** Arenga Palm Sugar Block, Arenga Palm Sugar Granule, Crystallinity, Microstructure, Morphology, Multilayer

### INTRODUCTION

The demand of arenga palm sugar granules is growing rapidly due to ease in use, handling, packaging, storage (Rao *et al.* 2010) [1] and contain more complete nutrition than can sugar (Bahr *et al.* 2008) [2]. One of the important nutrients of palm sugar granules is varying mineral. The EDX analysis shows that the mineral content of arenga palm sugar consists of potassium (2%), sodium (0.05%), magnesium (0.04%), calcium (0.01%), iron (0.11%), copper (0.75%), Zn (0.46%), manganese (0.08%) dan chromium (0.11). Because of these advantages, palm sugar granules become popular and demanded by developed countries. Farmers produce palm sugar granules by heating and evaporating sap up to  $120 \pm 2$  °C, then it cooled and stirred as sheared and pressed up into granules (called as wet granulation method). The daily

farmer productivity is around 6.5 kg/farmer. These small production is a result of the difficulty to collect large amount of sap, a relatively small amount of sap, a low ability of production skill owned by farmers and only a few farmers who are interested in processing palm sugar granules. For these reasons, if the palm sugar granules is produced by dry granulation method that started from palm sugar blocks, the above difficulties are expected to be overcome. It is easier to collect raw material in form of the palm sugar block than juice form and the production can be conducted on a larger production scale.

Some research about palm sugar has been conducted such as by Rao *et al.* (2010) [1] who studied the effect of moisture content on the glass transition temperature ( $T_g$ ) and sticky point temperature ( $T_{sc}$ ) on sugar cane, palm sugar and palm sugar granules, Naknean *et al.* (2009) [3] which characterized palm sap during heating into syrup, Suwansri *et al.* (2009) [4] who conducted the crystallization of palm sugar syrup by adding can sugar, Amin *et al.* (2010) [5] which examined the antioxidant activity of a palm sugar-like flavor on Maillard reaction of sucrose and amino acids (arginine, asparagine and glutamine), and Naknean (2010) [6] who examined factors that cause browning and crystallization of sugar syrup and sugar blocks from palm sap. These authors still less focusing on dry granulation using palm sugar blocks. The purpose of this study is to determine the physico-chemical properties of the arenga palm sugar blocks and granules, to analyze process phenomenon of arenga palm sugar granules by dry granulation and to obtain the structure and morphology of arenga palm sugar granules.

## METHODS

### Material and Equipments

Materials used in this study was arenga palm sap produced by farmers from Cikoneng Village, Sobang Subdistrict, Lebak District, Banten Province of Indonesia, saturated salt solution for  $a_w$  conditioning which was  $\text{LiCl}_2$ ,  $\text{CH}_3\text{COOK}$ ,  $\text{MgCl}_2$ ,  $\text{K}_2\text{CO}_3$ ,  $\text{NaBr}$ ,  $\text{NaNO}_2$ ,  $\text{NaCl}$ ,  $\text{KCl}$ , dan  $\text{K}_2\text{SO}_4$ , and chemicals for analyzing. Equipments used were slicer, tray dryer, stopwatch, glass thermometer, incubator, glass desiccator,  $a_w$  meter, oven, *X-Ray diffractometer* (Shimadzu XRD-7000), *Scanning electron microscopy* (SEM Zeiss Evo 50) and FTIR (ABB FTIR AB3000).

### Production of Arenga Palm Sugar Granules by Wet and Dry Granulation

Fresh palm sap in bamboo tube was poured, then was measured for its alkalinity and its solid content. It was then heated and evaporated in a open pan evaporator using firewood. Palm sap of 65 litres was heated around 97 °C up to  $120 \pm 2$  °C for about 3.5 hours. It was stirred occasionally during heating. The heating process then stopped and the thick palm sap was taken off the heating, after that it cooled for about 10 minutes while stirred, sheared and pressed up. When palm sugar was about to start forming granules, the stirring speed was accelerated. Arenga palm sugar granules from fresh palm sap was used as arenga palm sugar granules control (GAG-control). The producing of arenga palm sugar granules by dry granulation method was started with the preparation of the raw material of palm sugar block (GAC) from palm sap. The GAC process was same with the palm sugar granules by wet method but the heating was done up to  $118 \pm 2$  °C. Furthermore, the hot thick juice was poured on a wooden mold. It was settled in around 10 minutes until it become solid. After that, palm sugar block was removed from the molds and wrapped with sallaca leaves.

The next stage was dry granulation steps. First, GAC was weighed, then sliced by using a slicer (slicer) for about of 2-3 mm thickness. Next, palm sugar was placed on and spread thinly on shelves of 25 x 30 cm with a thickness of about 3 cm. Then it was heated in a dryer rack using air heated to a temperature of 70, 80 and 90 °C. Dry air flow rate of

2 liters per minute, inlet air RH of about 15-17.5% and exhaust air RH of about 35%. Every 5 minutes, the heated material was stirred, observed its physical changes and samples was taken until 45<sup>th</sup> minute. The arenga palm sugar granule from palm sugar block was called as GAG.

### Physico-Chemical Analysis

Physico-chemical analysis for arenga palm sugar blocks and granules consists of moisture content and insoluble material content using SNI method, ash content and total acid using AOAC 2000 method, reducing sugar and sucrose content using Luff Schoorl and DNS method.

### Structure Analysis

GAC and GAG microstructure were qualitatively analyzed using XRD-7000 X-Ray Diffractometer (Shimadzu XRD-7000) with x-ray Cu tube ( $\lambda = 0.154$  nm, energy input of 40 kV and 30 mA). Diffractogram was measured by *scan* speed of 2.0 degree per minute dan *pitch* sample of 0.02 degree. The GAC and GAG samples were put into aluminum sample-holder then pushed until compact using piece of glass to ensure the surface would be smooth and no hollow space. Then it was diffracted within interval 2-theta around 5° up to 60°.

### Morfology Analysis

Morphology and microstructure of arenga palm sugar blocks and granules were analyzed using “Scanning Electron Miroscopy” (SEM Zeis Evo 50, voltage speed of 10 kV). Samples were put and attached on metal stubs using double tape carbon and then coated using mixture of gold. Microscopic analysis was conducted in magnification of 50, 100 and 250 to obtain morphology structure, and sample size.

### FTIR Analysis

FTIR analysis was conducted to examine the functional groups on palm sugar block and granules. FTIR spectra were measured using a FTIR spectrometer (ABB FTIR MB3000). FTIR spectra were recorded in transmittance mode from 4000  $\text{cm}^{-1}$  to 800  $\text{cm}^{-1}$ .

### Determination of Sorption Isotherms

Sorption isotherms curve was plotted using “Static Vapor System”. For  $a_w$  conditioning, nine desiccators was prepared and each filled with 500 ml of saturated salt solution such as  $\text{LiCl}_2$ ,  $\text{CH}_3\text{COOK}$ ,  $\text{MgCl}_2$ ,  $\text{K}_2\text{CO}_3$ ,  $\text{NaBr}$ ,  $\text{NaNO}_2$ ,  $\text{NaCl}$ ,  $\text{KCl}$ , dan  $\text{K}_2\text{SO}_4$ , with their  $a_w$  of 0.11, 0.22, 0.32, 0.44, 0.56, 0.64, 0.75, 0.84, dan 0.97 respectively. Sample was weighed of 2 g and put into desiccators then placed in the incubator with temperature of 30° C. Palm sugar blocks and palm sugar granules were environmentally observed until their balance state were achieved and then equilibrium moisture content were measured.

## RESULTS AND DISCUSSIONS

### Physico-Chemical Characteristics of Palm Sugar Blocks and Palm Sugar Granules

Characteristics of arenga palm sugar block (GAC) showed the moisture and ash content (10.1% db and 3.3% db) greater compared to the quality standards, the Indonesian National Standard (8.7% db and 2.2% db). Other characteristics such as reducing sugar content (3.2% db), insoluble materials (0.54% db) and sucrose content (92.5% db) met quality standards, the Indonesian National Standard (respectively of 12.0% db, 1.1% db and 72.0% db).

Characteristic of palm sugar granules (GAG and GAG-control) showed significant difference with GAC. The GAG at temperatures of 70, 80 and 90 °C and GAG-control showed that their water content of 3.1, 3.1, 2.8, and 3.1% db respectively, reducing sugar content of 3.1, 2.0, 3.1 and 3.3% db respectively, sucrose content of 90.4, 78.8, 91.5 and 90.9% db respectively. All characteristics of GAG and GAG-control met to standards palm sugar granules, the Indonesian National Standard SNI 01-3743-1995. Those GAG and GAG-control with their characteristics as mentioned above, could be safely stored for a long time, it could be more than 6 months. GAG produced by the heating temperature of 80 °C gave the lowest reducing sugar, however, it also gave the value of the lowest levels of sucrose as well. The highest sucrose levels indicated by GAG with a temperature of 90 °C.

The condition of the GAC moisture content higher than standard but it had a high levels of sucrose and a low level of reducing sugar, indicated that the GAC still could be stored in expected time by arranging a proper storage room RH. By the right set up of the room RH, the moisture content of the GAC would continue to decrease until it reaches the equilibrium moisture content (it was showed in the analysis of sorption isotherm in the subsequent discussion below). If the room's RH was not appropriate to storage condition needed, it tended to high damage. It was probably caused by the enzymatic hydrolysis during storage. Sucrose would be hydrolyzed into simple sugars (reducing sugars). Further reducing sugars tended to hinder of palm sugar block to form palm sugar granules (GAG). At the time of heating, reducing sugars interacted with proteins and Maillard reaction occurs to form a dark color (Naknean *et al.* 2009) [3] and aroma (Wai *et al.* 2005) [7].

#### Phenomenon of the Characteristics Changes of the Material During the Granulation Process

In the early stages until the 10<sup>th</sup> minute of dry granulation process, the material condition was similar to the starting material. In the 15<sup>th</sup> minute (for the temperature of 80 and 90 °C) and 20<sup>th</sup> minutes (for the temperature of 70 °C) the material began to dry on the surface but still looked a bit sticky. In the 25<sup>th</sup> minute until the 30<sup>th</sup> minute (for the temperature of 80 and 90 °C) and 30<sup>th</sup> minutes to 35<sup>th</sup> minutes (for the temperature of 70°C) the material was relatively non-sticky and easily broken into granules. In this condition, its moisture content ranged between 4.5-3.3% (Table 1). Temperature of 80 and 90 °C exhibited almost similar effect to the time of granulation process. So that the temperature of 80 °C would be more efficient than the other, and it candidated as the best one. Meanwhile, the temperature profile of the material during the granulation process tended to increase sharply until the 20<sup>th</sup> minute with the temperature of material reached of 60, 65 and 75°C for inlet air temperature of 70, 80 and 90°C respectively.

**Table 1: Physical Profile and Moisture Content of Palm Sugar Granules During Granulation Process with Air Inlet Temperature of 70 °C, 80 °C Dan 90 °C**

| Processing Time (minute) | Air Inlet Temperature (°C) |                         |                      |                         |                      |                         |
|--------------------------|----------------------------|-------------------------|----------------------|-------------------------|----------------------|-------------------------|
|                          | 70                         |                         | 80                   |                         | 90                   |                         |
|                          | Physical Appearance*       | Moisture Content (% db) | Physical Appearance* | Moisture Content (% db) | Physical Appearance* | Moisture Content (% db) |
| 0                        | 0                          | 9,52                    | 0                    | 9,52                    | 0                    | 9,52                    |
| 5                        | 0                          | 9,43                    | 0                    | 9,04                    | 0                    | 8,63                    |
| 10                       | 1, 2                       | 8,13                    | 1, 2                 | 8,31                    | 1, 2                 | 8,00                    |
| 15                       | 2                          | 6,71                    | 1,2,3                | 6,71                    | 1,2,3                | 6,51                    |
| 20                       | 2, 3                       | 6,05                    | 2, 3, 4              | 5,22                    | 2, 3, 4              | 5,45                    |
| 25                       | 2, 3, 4, 5                 | 4,69                    | 2, 3, 4, 5           | 4,23                    | 2, 3, 4              | 4,11                    |

Table 1: Contd.,

|    |            |      |         |      |            |      |
|----|------------|------|---------|------|------------|------|
| 30 | 2, 3, 4, 5 | 3,86 | 3, 4, 5 | 3,38 | 2, 3, 4,   | 2,66 |
| 35 | 3, 4, 5    | 3,61 | 3, 4, 5 | 2,99 | 3, 4, 5, 6 | 2,06 |
| 40 | 4, 5, 6    | 2,69 | 6       | 2,54 | 6,7        | 1,97 |
| 45 | 5, 6, 7    | 2,63 | 7       | 2,31 | 7          |      |
| 50 | 6, 7       | 2,42 |         |      |            |      |

**\*Physical appearance:**

- |                                       |                   |
|---------------------------------------|-------------------|
| 0-Similar to starting material        | 4-Easily broken   |
| 1-Begins to dry on the surface        | 5-Flow and hablur |
| 2-Starting to break, but a bit sticky | 6-Starting to dry |
| 3-Easily broken, non-sticky           | 7-Dry condition   |

In contrast with the material temperature, moisture content of materials continued to decline rapidly until the 20<sup>th</sup> minute down to about 5-4 % db (Figure 1b and Table 1). This condition informed that granules/particles formation was good when the moisture content coincides multilayer water (3.8%) according to sorpsi isotherm showed at Figure 6 below. In the 40<sup>th</sup> minute the temperature of the material increased again with a relatively small value. These conditions indicated the material almost reached the monolayer water which was difficult to be evaporated, so that thermal energy was used to increase the temperature of the material. In the 45<sup>th</sup> minute temperature looked slightly increase and water content tend to decrease nearly 2%, that the moisture content closed to the monolayer water (about 2%).

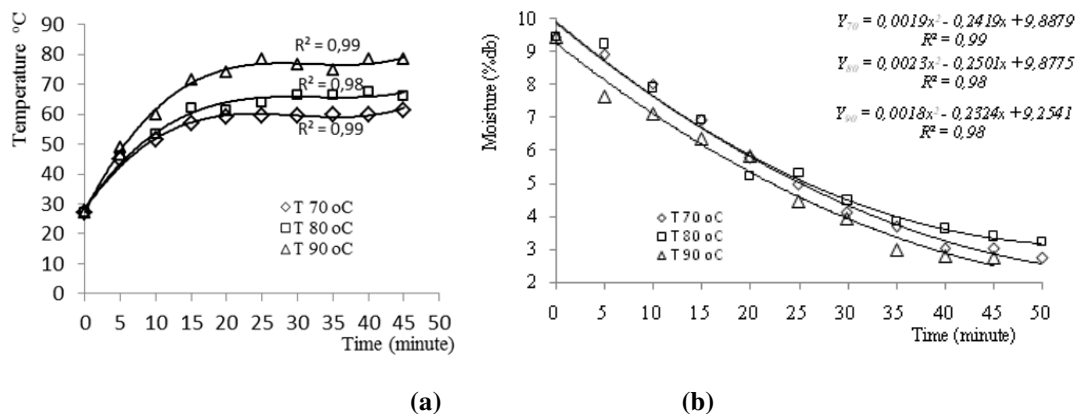
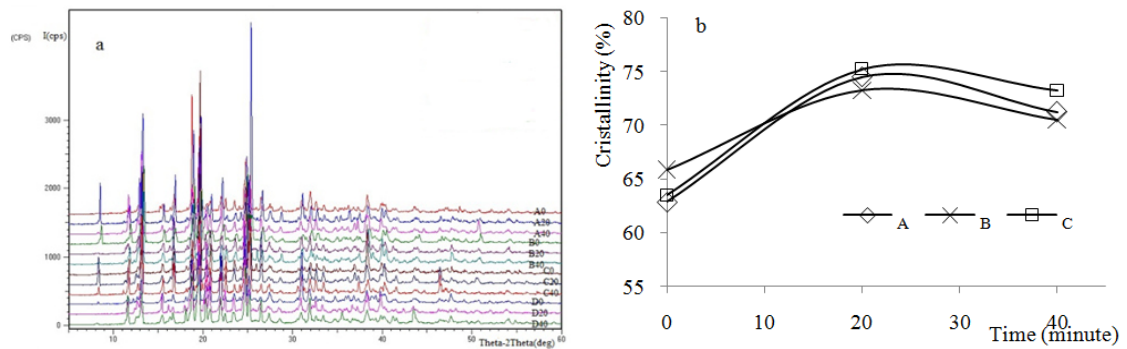


Figure 1: Profile of (a) Temperature and (b) Moisture Content of Material in Granulation Process by Using Hot Air Temperature of 70, 80 Dan 90 °C

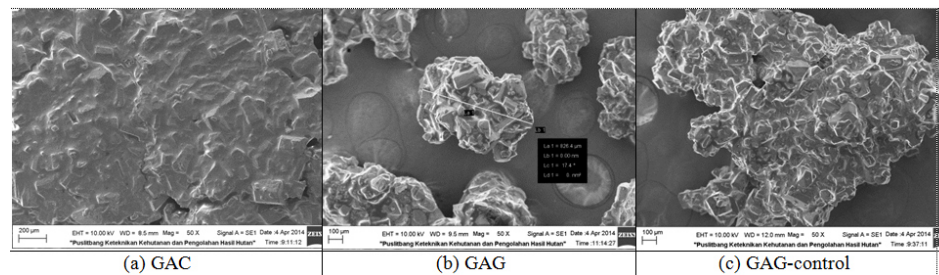
### Structure and Morphology

Diffractiongram of GAC and GAG (Figure 2a) showed that the GAC and GAG's X-ray diffraction patterns had relatively same structure. The differences of their diffraktograms were focused on their intensity. The changes that occurred in the granulation process were that GAC's crystallinity increased for all temperature of 70, 80 or 90 °C. These conditions informed that amorphous phase component had high reactivity to transform into crystalline phase (crystallinity level). The temperature of 80 °C exhibited the higher cristalinity compared to the other. Figure 2a (A, B, C) showed some small peak grew compared to Figure 2a (D) or increasing the intensity of the existing peaks, so that the crystallinity increased.



**Figure 2: Profile of (a) Diffractogram and (b) Crystallinity of GAG by Dry Granulation at Temperature of (A) 70 °C, (B) 80 °C and (C) 90 °C. Number follow A, B, C and D are Time in Minute**

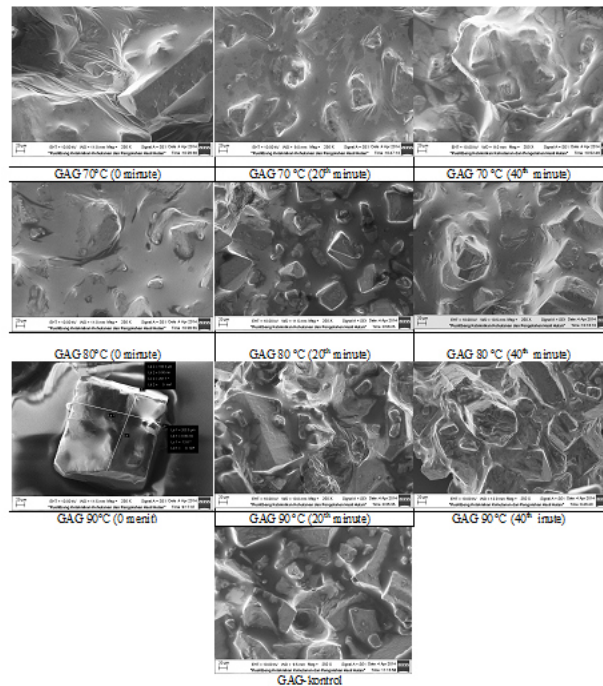
All GAC and GAG showed the similar diffractogram, so that their structure constructed by the relatively same components or compounds. The XRD analysis showed that crystallinity increased during the granulation process until the 20<sup>th</sup> minute but it declined in the 40<sup>th</sup> minute. At the beginning of the process, material had a crystallinity of about 63-66%, after 20 minutes it increased to about 75% and then it decreased again at the end of the process into about 72% (Figure 2b). According to Rao *et al.* (2010) [1] amorphous phase was formed during cooling of the melt material or rapid discharged water during dehydration. Fenema (1985) [8] stated that the sugar's hydroxyl groups played a greater role in the occurrence of hydrogen bonds with the surrounding water. Thus, sugar crystallization occurred due to loss of water within sufficient time, so that the atom molecule had enough time to arrange itself to form crystal for the discharge of water. Furthermore, according to Harnkarnsujarit and Charoenrein (2011) [9], sugar that formed hydrated crystals (dihydrates trehalose and raffinose with tri, tetra or penta hydrate) retained high amounts of water, while the anhydrous crystalline of sugar (sucrose and lactose) released all the water after crystallization.



**Figure 3: Morphology Profile of (a) GAC, (b) GAG Processed at 80 °C and (c) GAG-Control**

The SEM analysis of GAC, GAG and GAG-kontrol (Figure 3), showed that GAC consisted of sucrose crystal aggregation binded by dough-like binder. Palm sugar granule of GAG and GAG-control showed morphologically differ in size. A large aggregations on GAC as GAG's material was converted into granules through a size reduction and heating in order to obtain a stable granules. Morphology of GAG or GAG-control was not much different from the GAC, it showed that the granulation process did not change the basic morphological structure of the GAC. Dry granulation changed the size and shape became smaller in more stable condition. Figure 3 shows the GAG and GAG-control had a nearly spherical to irregular shape.



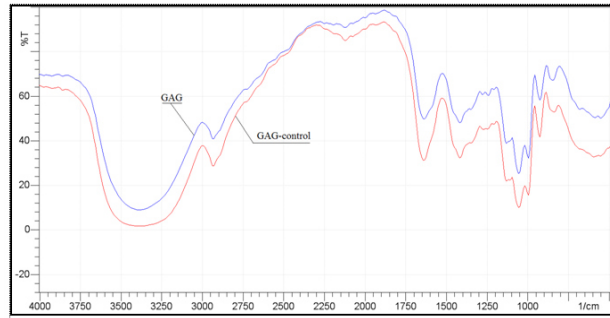


**Figure 4: Profile of Morphology of GAG at Temperature of 70, 80 Dan 90 °C and GAG-Control**

Morphology of GAG and GAG-control consisted of octahedron crystal particles were wrapped by a layer of binder. The binder layer was suspected as part of the fine crystal, the hygroscopic amorphous part, water and other components then they mixed to form dough-like binder. Morphology of GAG and GAG-control seemed almost similar at every stage, however, by the time increased, it seemed to increase to become a stronger bond. This was indicated by the protruding part of crystal growing shrunken part due to the drying process. The surface morphology of dry granular particles of palm sugar affected the physical and functional properties of palm sugar granules. Granulation process produced granules consisting of a group of individual crystalline particles, which bound or covered by a bonding material such as amorphous materials and water (Figure 4). Effect of temperature at 70, 80 and 90 °C resulted in the shape and size of the granules was not much different from its morphology. Particle surface appeared rougher due to the drying process that induced the amorphous. The structure of hollow/porous that were formed during the process gave the soluble and water migration during hydration. The existence of an amorphous phase in the GAG indicated the product was hydrophilic material, therefore packaging issues would significantly determine the stability of the product. Packaging should have a good barrier properties against water vapor.

#### FTIR Analysis

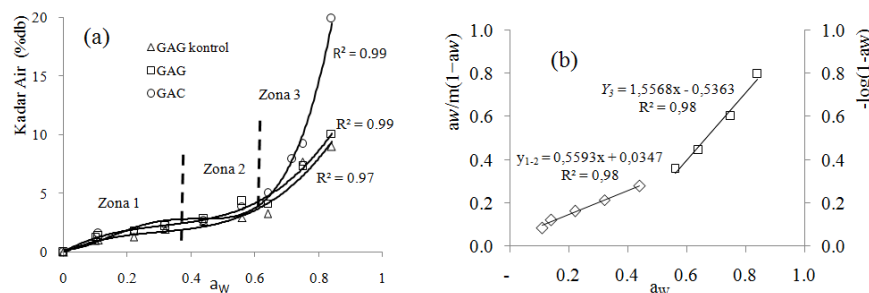
Functional groups of palm sugar granules using the dry granulation (GAG) compared with palm sugar granules by wet granulation (GAG-control), showed no significant difference. The results of FTIR analysis (Figure 5) showed the same shape of their transmittance curves. The most clear difference was the transmittance intensity of GAG higher than the GAG-control.



**Figure 5: Transmittance of FTIR Analysis of GAG and GAG-Control**

The FTIR spectra confirmed the basic structure GAG and GAG-control. The strongest of polar covalent bond was the carbonyl group (C=O) in the wavenumber (frequency) between 1700 to 1600  $\text{cm}^{-1}$  (Volland 1999) [10]. Hydroxyl group (OH) of hydrogen bonds was within frequency 3600-3000  $\text{cm}^{-1}$  (Koay et al. 2011 [11]; Ma *et al.* 2013 [12]; UI-Islam *et al.* 2013 [13]; Suvakanta *et al.* 2014 [14]) of which GAG and GAG-control were centralized at a frequency of 3387  $\text{cm}^{-1}$ . Hydroxyl groups of hydrogen bonding, contribute to the structure of the sugar compound (Suvakanta *et al.* 2014 [14]) as the hydroxyl sugar. Spectra that appeared at frequency of 2939  $\text{cm}^{-1}$  were the CH bond stretch (UI-Islam *et al.* 2013 [13]; Ma *et al.* 2013 [12]; Suvakanta *et al.* 2014 [14]; Wang *et al.* 2014 [15]).

The existence of CH spectrum together with the presence of the carbonyl (C=O) bond, was an aldehydes group (Volland 1999) [10] of sugar samples. Meanwhile, according to Liet *et al.* (2014) [16], the emergence of the peak at a frequency of 1643  $\text{cm}^{-1}$  showed the HOH functional groups which indicated that the GAG and GAG-control absorb water (hydrophilic). The peak at 1420  $\text{cm}^{-1}$  indicated the presence of the existence CH-bonds bending, whose presence together with stretching carbonyl (C=O) indicated the presence of aromatic groups (Volland 1999) [10]. At frequency of 1273  $\text{cm}^{-1}$  appeared spectrum indicates a stretching CO bond, whose existence with the carbonyl group (C=O) and hydroxyl (OH) indicated the presence of carboxylic acids (Volland 1999 [9]). At frequencies between 1780-1620  $\text{cm}^{-1}$  indicated the presence of carboxyl groups (COOH) (Koay 2011 [11]; UI-Islam 2013 [13]; Rockwell 2014 [17]).



**Figure 6: Equilibrium Moisture Content Curve of (a) GAC, GAG and GAG-Control and (b) BET Regression Curve Example for GAG**

The wavenumbers (frequency) between 1780-1620  $\text{cm}^{-1}$  indicated the presence carboxyl group (COOH) (Koay 2011 [11]; UI-Islam 2013 [13]). At the frequency of 1273  $\text{cm}^{-1}$  indicated the presence of functional groups CO (ester) and at 1134  $\text{cm}^{-1}$  indicated COC functional groups while at 1057  $\text{cm}^{-1}$  was the functional group of polymer backbone chains C-C of GAG (Rockwell 2014) [17]. The peak at 1134  $\text{cm}^{-1}$  and 1057  $\text{cm}^{-1}$  indicated the presence of galactans and arabinogalactan (Kacurakova 2000) [18] and was also suspected as a pyranose ring, while the peak at 849  $\text{cm}^{-1}$  wave as an indication of the presence of mannose (Wang *et al.* 2014) [15].



### Sorption Isotherms

The behavior of GAG and GAC water sorption (equilibrium moisture content) at a certain level of  $a_w$  are showed in Figure 6a. The relationship between equilibrium moisture content and water activity ( $a_w$ ) produced J-like curve (J-type isotherm) which was characteristic of the products that contain high sugar and little non-sugar fraction, which was a soluble molecule and containing little polymeric materials (Fennema 1985) [8]. From the figure, the equilibrium water content increased with increasing of  $a_w$  and sharper in  $a_w$  above 0.60. According to Fennema (1985) [8], the minimum reaction rate was typically found at the boundary between zone 1 and zone 2 sorption isotherm of the system. The boundary zones 1 and 2 was a monolayer of water content. Monolayer water content gives the first allegation as water levels provide maximum stability. The monolayer water layer had a low molecular mobility, not capabled of dissolving solute and played a role in chemical reactions (Saavedra-Leos, 2014) [19].

Determination of water monolayer value was done by using the regression curve Figure 6b and equation 1, 2, while the determination of multilayer was done using equation 1 and equation 3 which was developed by Brunauer, Emmett and Teller (BET) as BET equation such as the following:

for zone 1 and 2

$$\frac{a_w}{m(1-a_w)} = \frac{1}{m_1 c} + \frac{c-1}{m_1 c} a_w \quad (1)$$

$$m_1 = \frac{1}{Y_{x=0} + slope} \quad (2)$$

for zone 3

$$-\log(1 - a_w) = a + b a_w \quad (3)$$

where  $m$  was the equilibrium moisture content at  $a_w$  levels,  $m_1$  was the monolayer moisture content and  $c$  was a constant of energy (Fennema, 1985) [8].

By plotting the data of each curve in Figure 6a with equations 1 and 2 above, the obtained regression equation of BET that fit of each zone for each palm sugar granules and blocks were stated as the following:

$$\text{GAG} \quad Y_{1-2} = 0.5593 x + 0.0347, R^2 = 0.98 \text{ dan } Y_3 = 1.5568 x + 0.3563, R^2 = 0.98$$

$$\text{GAG-kontrol} \quad Y_{1-2} = 0.7378 x + 0.0269, R^2 = 0.95 \text{ dan } Y_3 = 1.5568 x + 0.5363, R^2 = 0.98$$

$$\text{GAC} \quad Y_{1-2} = 0.6770 x + 0.0365, R^2 = 0.99 \text{ dan } Y_3 = 1.5462 x + 0.5352, R^2 = 0.97$$

At equilibrium GAG, if  $\frac{a_w}{m(1-a_w)}$  as  $Y_{1-2}$  (Y at zone 1 and 2) and  $a_w$  as  $x$ , by using regression equation from Figure 6b, when  $a_w = 0$ , value of  $Y_{1-2, x=0} = 0.0347$  and by using equation 2 then obtained  $m_1 = 1.68\%$  as monolayer water which fit the condition of  $a_w = 0.20$ . Then with  $-\log(1 - a_w)$  as  $Y_3$  (Y at zone 3), the intercepts of the regression line in equation 1 (or regression equation  $Y_{1-2}$ ) and regression line from equation 3 (or regression equation  $Y_3$ ) was a moisture content limit of multilayer water. Those two regression lines intercepted at  $a_w = 0.57$  with equilibrium moisture content of 3.78% as multilayer water or the highest limit of water fraction of zone 2. That multilayer water was on a solid surface and condensed on a capillary which had a role in biochemical reaction and functioned as solvent for molecular weight solutes with low molecular weight (Fennema 1985) [8]. In zone 3 with  $a_w$  values above 0.57, the water molecules form a

multilayer located in the space between the free molecule or forming liquid phase. Water was available as solvent and the growth media of microorganisms. At a likely higher  $a_w$  or above 0.57, water adsorption was increased by the higher value of  $a_w$ . Increased moisture content was due to the availability of functional group (active group) as glucose and fructose as reducing sugar. The surface of the sugar which had hydroxyl groups (OH) were somewhat big, because the water molecule group adsorbed physically and interact with hydroxyl groups form hydrogen bonds to further dissolve of sugar and other sugar polymers (Fenema 1985) [8].

As showed in Figure 6a, the behavior of water sorption in the GAG had the same curve shape with GAG-control and palm sugar blocks (GAC), which was of J curve. In the same way, the moisture content of monolayer and multilayer for GAC and GAG-control were obtained and showed in Table 2. Table 2 showed that the GAG-control had monolayer moisture content of 1.3% on  $a_w = 0.22$  and multilayer moisture content of 3.9% on  $a_w = 0.64$ . The GAG palm sugar granules produced monolayer moisture content lower (1.68%) compared to monolayer moisture content of palm sugar blocks (1.79%) and so was the multilayer water of palm sugar granules also smaller (3.78%) compared with palm sugar blocks (5.08%). The condition of equilibrium moisture content in the form of monolayer of palm sugar blocks and palm sugar granules were at a very similar water activity, between  $a_w = 0.19$  to 0.21, whereas GAG's multilayer value had lower  $a_w$  (0.57) among the others (0.64 - 0.66). From the sorption isotherms analysis and granulation process, it could be concluded that the best granulation process occurs when water reached a multilayer water condition.

**Table 2: Monolayer and Multilayer Water at Each  $A_w$  of Palm Sugar Blocks and Palm Sugar Granules**

| Palm Sugar Classification | Monolayer Water (Zone 1 and 2) |       | Multilayer Water (Zone 3)    |       |
|---------------------------|--------------------------------|-------|------------------------------|-------|
|                           | Moisture Content ( $m_1$ ) %   | $a_w$ | Moisture Content ( $m_2$ ) % | $a_w$ |
| GAG                       | 1.68                           | 0.20  | 3.77                         | 0.57  |
| GAG-control               | 1.27                           | 0.21  | 3.87                         | 0.64  |
| GAC                       | 1.79                           | 0.19  | 5.08                         | 0.66  |

## CONCLUSIONS

This study has revealed that Arenga palm sugar block (GAC) can be transformed successfully into arenga palm sugar granule (GAG) by dry granulation method. Temperature and time had significant effect on dry granulation process. The best granulation occur when the moisture content of processed material reach around multilayer level. The temperature of 70, 80 and 90°C give different effect on crystallinity but tend to similar effect on morphology of GAG. Temperature of 80 °C is suggested to be a better than other. Although the crystallinity of GAG is different with GAC, their morphology and microstructure are not different dramatically The dry granulation has the same effect on functional group compare with wet granulation. The dry granulation at 80 °C is recommended for future work to produce palam granule.

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