Kinetics Physical Properties of Coconut Sugar Solution, During Processing Palm Sugar Using Pan Evaporator and Rotating Crystallizer

Destia Catur Rini1,* Sri Rahayoe1 Arifin Dwi Saputro1 Joko Nugroho Wahyu K1

1Department of Agricultural and Biosystem, Faculty of Agricultural Technology *Corresponding author. Email: destia.catur.rini@mail.ugm.ac.id

ABSTRACT
Palm sugar is sugar from coconut sap in the form of crystals. The producers of palm sugar are mostly home industries that process sugar in a conventional way, using wood-fired stoves for thickening sap and the crystallization process is done manually. Improvements to the conventional method carried out in this research were the use of a pan evaporator and a rotary crystallizer for the processing of palm sugar from coconut sugar raw materials. This research aims to analyze the kinetics of the physical properties of coconut sugar solution during the processing of palm sugar.

The raw material for making palm sugar is coconut sugar which is dissolved in water in a ratio of 1 kg: 500 ml. The sugar solution was thickened using a pan evaporator until a supersaturated sugar solution was obtained and continued with the crystallization process using a rotary crystallizer. As a comparison, the crystallization process was also carried out manually. During the thickening and crystallization process, physical properties of the cave solution were measured, including temperature, brix, and density at certain time intervals. The data on changes in physical properties were analyzed using Avrami kinetic models and first order kinetics.

The results showed that the Avrami model can be used properly to predict changes in the physical properties of sugar solution temperature, brix, and density during the evaporation and crystallization processes. While the first-order kinetic model can be used to explain the phenomenon of temperature changes during the crystallization process. Avrami constant (k) changes in temperature, brix, and density in the evaporation process, respectively 0.004±2x10^-3 - 0.007±2x10^-3 °C/min; 1.07x10^-3±0.3x10^-3 - 1.18x10^-3±1x10^-4 %/minute; 2.08x10^-4±4x10^-5 - 2.73x10^-4±4x10^-5 (kg/m³)/minute and the nA values ranged from 1.77 to 1.94, while during the crystallization process the temperature and density constants ranged from 0.109±0.01 - 0.116±0.01 °C/min; 1.67x10^-1±0.5x10^-1 - 1.7x10^-1±4x10^-4 (kg/m³)/min.

The results of the physical characteristics of palm sugar showed water content 1.38±0.6 - 1.79±0.3%, yield 78.42±3.6 - 79.13±3.4%, fineness modulus 3.95±0.36 - 4.53±0.67, particle diameter 0.065±0.01 - 0.101±0.04 inches, and the color was brownish-yellow.

Keywords: Palm sugar, evaporation, crystallization, constant, characteristics.

1. INTRODUCTION
Palm sugar is a sugar product in the form of small granular powder (granules). The consumption of sugar as a sweetener continues to expand with significant increase, both for household and industrial consumption. The demands for palm sugar exports during January-March 2020 reached 311 tons with an export value of Rp. 19.27 billion, with most of these palm sugar exports were used to meet the needs of the United States market and the rest were exported to Germany, England, Serbia, Malaysia, Australia, Hong Kong and Turkey [1]. The advantage of palm sugar compared to other sweeteners are: it can be dissolved easily in the water, it’s storable, easy to pack and it has been claimed to have a lot of health benefits such as a low glycemic index ranging from 35-42, antioxidants,
vitamins and minerals thiamin, riboflavin, nicotinic acid, and ascorbic acid [2].

In Indonesia, palm sugar is still mostly made conventionally by household craftsmen in small to medium industries. The conventional process covers the stages of the cooking process of sap or coconut sugar solution using a wood-fired stove, so it causes a few negative effects such as: smoke contamination, uncontrolled temperature, and longer cooking process which takes a lot of time. The next stage is the crystallization process with manual stirring method. The conventional production process of palm sugar results in variations of granules and decreased productivity, thus some mechanization methods were needed. We needed to conduct research on the process of palm sugar production to improve the quality of palm sugar that still follows the Indonesian National Standard and exportation clauses and rules in Indonesia.

This research was conducted using a mechanical device such as a pan evaporator and a rotary crystallizer. The pan evaporator is a direct type heater with a heating source of fire, designed with a hygienic furnace, made of stainless steel coated with glass wool as a heat insulator so the heat is evenly distributed to minimize the heat loss. The pan evaporator was equipped with a chimney as a smoke outlet from the combustion to minimize pollution in the making of palm sugar process. Meanwhile, the rotary crystallizer is designed with two static stirrers so that the stirring of the sugar solution is controlled. To ensure that the use of palm sugar could produce a product with good quality characteristics, it was necessary to obtain knowledge about the physical and chemical properties of the sugar. Therefore, the purpose of this research was to overcome the problem of conventional palm sugar processing in terms of the physical properties of palm sugar.

2. MATERIAL AND METHODS

This research was conducted in June 2020-April 2021, in the Food and Postharvest Engineering laboratory, Department of Agricultural and Biosystem Engineering, Faculty of Agricultural Technology, Gadjah Mada University.

2.1. Material and Tools

The materials used in this study were aquadest and molded sugar obtained from Kokap, Kulon Progo Regency, Special Region of Yogyakarta Province. The coconut sugar was dissolved in the aquadest in a ratio of 1 kg: 500 ml. The characteristics of coconut sugar can be seen in Table 1.

<table>
<thead>
<tr>
<th>Condition sugar</th>
<th>description</th>
</tr>
</thead>
<tbody>
<tr>
<td>mass of piece sugar</td>
<td>125-150 gr</td>
</tr>
<tr>
<td>pH</td>
<td>5.2-5.51</td>
</tr>
<tr>
<td>Density</td>
<td>1.2-1.5 kg/m³</td>
</tr>
<tr>
<td>color</td>
<td>brown-yellow</td>
</tr>
</tbody>
</table>

This research used the main tools such as a pan evaporator and rotary crystallizer, while other supporting tools are a pH meter, a refractometer, a thermocouple, a thermohygrometer, a color meter, a thermo gun, and a scale.

2.2. Sample preparation

This research used different tools method the palm sugar produced are named in Table 2.

<table>
<thead>
<tr>
<th>Type tools</th>
<th>Product</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pan Evaporator</td>
<td>GSPE</td>
</tr>
<tr>
<td>Rotating Crystallizer</td>
<td>GSKP</td>
</tr>
</tbody>
</table>

2.3. Palm sugar processing

The making of palm sugar process consists of several stages which include dissolved coconut sugar in aquadest in a ratio of 1 kg: 500 ml, evaporation, crystallization, drying, sifting and packaging.

2.3.1. Evaporation

We cooked the sugar solution using a pan evaporator. When the sugar solution boils, we stirred it manually. Measurements of temperature, density, ambient humidity, and brix were carried out every 3 minutes starting from t = 0. The temperature measurement was carried out at several points of the evaporator. The brix observation was carried out by taking 1-2 drops of a sample of sugar solution placed in a refractometer, while the density measurement was carried out by taking samples up to a volume of 5 ml. The evaporation process was stopped when the sugar solution was oversaturated and the water had evaporated to at least 80%. It was characterized by the alteration of saturated sugar solution (viscous) into a supersaturated sugar solution.

2.3.2. Crystallization

The crystallization process consisted of the formation process of the supersaturation condition, nucleation or formation of crystal nuclei, and crystal growth and recrystallization. With a gradual decrease in temperature and the stirring of the solution, sugar crystals could be formed. In this research, the stirring process was done manually and mechanically. The manual stirring method was done by attaching the material to the wall of the pan, then the material was
stirred manually until the crystal granules were formed. In the mechanical stirring method, we used a rotary crystallizer by transferring the materials from the evaporator pan to the crystallizer pan. During the crystallization process, we also calculated and measured the values of changes in temperature, brix, density, and environmental humidity. When sugar granules had been formed, the crystallization process was stopped.

2.3.3. Drying

The palm sugar obtained from the crystallization process was still in a wet condition, so it needed to be dried first so that the sugar would not stick together. We did the drying process using a cabinet dryer at a temperature of 50 °C for 6 hours. During the drying process, the palm sugar was turned over every 1 hour so that the sugar could dry evenly and perfectly.

2.3.4. Sieve tyler and Packaging

The sifting process was done to uniformize the particle size of palm sugar. Before starting the sugar sifting, the palm sugar was weighed, then we prepared the tyler sieve. The palm sugar was sieved using a Tyler sieve with sieves sized 8, 14, 30, 40, 50, 100 for 10 minutes. Then the palm sugar was packaged using a standing pouch and then we added silica gel to maintain the water content in the package.

2.4. Analytical methods

Changes in temperature, brix and density could be modeled using the Avrami equation. The Avrami equation analyzes the rate of increase in temperature, brix and density during the evaporation and crystallization processes. The Avrami equation is modeled as follows:

\[ \varphi(t) = 1 - \exp(-k \cdot t^{nA}) \]  

where:

- \( \varphi \) = Avrami’s model changes to time
- \( k \) = Coefficient rate of change
- \( nA \) = Constant Avrami reference value 0.5-4

2.4.1. Temperature

The temperature measurement was done during the evaporation and crystallization process using a thermo gun or a calibrated thermocouple. We did the temperature measurement by firing a thermo gun directly at the temperature that would be calculated later. The shooting was done by pressing the measuring trigger button for 3 seconds and then releasing it after. Measuring the temperature with a thermocouple began with placing the sensor cable to the temperature point then pressing the power button (on). The temperature measurement was carried out every three minutes during the evaporation and crystallization process. Changes in temperature during the evaporation process were analyzed using the Avrami equation to obtain constant values (k) and nA. Modification of the Avrami equation was used to calculate the prediction temperature during the evaporation process with the following equation:

\[ \frac{t_e - t_0}{t_e - t_0} \approx \frac{t - t_0}{t_e - t_0} \]  

\[ T_t - T_0 = \left( 1 - \exp(-k_b \cdot t^{nA}) \right) \cdot (T_0 - T_e) + T_0 \]  

During the crystallization process, the temperature of the material decreased from high temperature to ambient temperature. In first-order kinetics, the speed of a process is directly proportional to the length of time. The shape of the curve in first-order kinetics is linear. Figures and kinetic equations of order 1 can be modeled with the following equation:

\[ \frac{dx}{dt} = \pm k \]  

\[ \frac{x}{t} \]  

\[ x \]  

\[ dx = \int_0^t \pm k \cdot dt \]  

\[ \ln \frac{x}{t} - \ln x_0 = \pm k \cdot t \]  

\[ \ln \left( \frac{x}{t} \right) = \pm k \cdot t \]  

2.4.2. Brix

The Brix measurement was done with two repetitions using a refractometer. However, a refractometer is only able to measure the degree of brix with a scale of 0-85% so that the brix measurement was done by two methods. The first method that we did was that the sugar solution was dripped on the refractometer and when we pressed the start button, the brix degree value would appear. Data collection with this method is used if the brix degree value is between 0-85%. We used the second method when the brix degree value was >85%, data collection was done by diluting the palm sugar solution with a ratio of the volume of the sample sugar solution and aquadest of 2:3. Then the result of solution dilution was dripped into the refractometer and then we pressed the start button so that we could read the brix value. That value was not the real brix, so it was necessary to find the actual brix value first. The actual brix value can be calculated using the following equation:

\[ \text{Brix sebenarnya(%)} = \frac{m_s - m_0}{m_0} \times \text{brix terbaca (℅)} \]  

Equation 1 can be assumed for brix change kinetics condition and that condition can be modified on Avrami model to analyze the brix change kinetics. The modification of the Avrami equation was done to determine the prediction brix which could be written with the following equation:

\[ \frac{B_t - B_0}{B_e - B_0} \approx \frac{B_t - B_0}{B_e - B_0} \]  

\[ B_t - B_0 = \left( 1 - \exp(-k_b \cdot t^{nA}) \right) \cdot (B_0 - B_e) \]
$B_2 = \left[ 1 - (\exp(-k_c t^{nA})) \right] \cdot (B_0 - B_2) + B_0 \quad (13)$

### 2.4.3. Density

The density measurement was done by weighing a container whose volume was known to be 5 ml, then we filled it with a palm sugar sample and we weighed the total weight. The density can be calculated with the following equation:

$$m = m_1 - m_2 \quad (14)$$

$$\rho = \frac{m}{V} \cdot \frac{1 \, \text{kg}}{10^3 \, \text{g} \cdot \text{m}^3} \quad (15)$$

Where:

- $\rho$ = Density (kg/m$^3$)
- $m_1$ = cup mass (g)
- $m_2$ = cup mass + sample mass (g)
- $m$ = sample mass (g)
- $V$ = sample volume (m$^3$)

Changes in density in the palm sugar production process can be modeled with the Avrami equation to obtain the values of $k$ and $n_A$. The Avrami equation was modified according to the conditions of the materials at the beginning of the process, during the process, and at the end of the process. Modification of Equation 1 was done to determine the prediction of Equation 1. was done to determine the prediction density which can be written with the following equation:

$$\rho_e - \rho_0 = \left( \exp(k_p t^{nA}) \right) - 1$$

$$\rho_t - \rho_e = \left( \exp(k_p t^{nA}) \right) - 1 \cdot \left( \rho_e - \rho_0 \right)$$

$$\rho_t = \left( \exp(k_p t^{nA}) \right) - 1 \cdot \left( \rho_e - \rho_0 \right) + \rho_0$$

### 2.4.4. Particle size

The distribution of sugar particles was determined by measuring the mass of granulated sugar left in each mesh, then the mass was used as a mass fraction by using the following equation:

$$\% \text{ Mass fraction} = \frac{m_i}{100} \quad (20)$$

$$\text{Oversize Xi} \, \% = \frac{\sum m_i}{\sum m} \times 100\% \quad (21)$$

Where:

- $m_i$ = mass of material retained per mesh (gr)
- $\Sigma m_i$ = total mass of material retained per mesh (gr)

The mass in the equation was the mass in the 8-100 mesh, and we didn’t have to use the pan. To determine the cumulative oversize mass fraction, we can use the formula in Table 3.

### Table 3. Determination of the cumulative Mass Oversize Fraction

<table>
<thead>
<tr>
<th>No Mesh</th>
<th>Holec size (mm)</th>
<th>Massa of material retained (mi)</th>
<th>Oversize Xi (%)</th>
<th>Oversize cumulative (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>2.8393</td>
<td>m1</td>
<td>X1</td>
<td>X1</td>
</tr>
<tr>
<td>14</td>
<td>1.8796</td>
<td>m2</td>
<td>X2</td>
<td>X1+X2</td>
</tr>
<tr>
<td>30</td>
<td>0.9995</td>
<td>m3</td>
<td>X3</td>
<td>X1+X2+X3</td>
</tr>
<tr>
<td>40</td>
<td>0.5093</td>
<td>m4</td>
<td>X4</td>
<td>X1+X2+X3+X4</td>
</tr>
</tbody>
</table>

The degree of fineness (Fineness Modulus) of palm sugar can be calculated with the following equation:

$$\text{FM} = 100 \frac{\text{oversize cumulative}}{\text{oversize Xi}}$$

After obtaining the fineness modulus, the next step was to calculate the particle diameter. The relationship between the average diameter and the degree of fineness can be written with the following equation:

$$D_i = \frac{0.0041 \cdot (FM)}{2}$$

Where:

- $D_i$ = particle diameter (inch)
- $D_{pi}$ = particle diameter $i$
- $\text{oversize cumulative}$

### 2.4.5. Color

The measurement of palm sugar’s color using a colorimeter. The measurement was done by firing a colorimeter at the sugar sample so that it would show some color indicators named $L^*$, $a^*$, $b^*$. The color measurements of palm sugar were repeated 3 times.

### 2.4.6. Moisture content

The measurement of the water content was carried out by weighing the cup using an analytical balance, then the palm sugar was put in a 2-3 grams cup and then we weighed it. This was done with three replications, then the sample was oven-dried at 1050°C for 3 hours to achieve the constant weight. The cooled sample and cup were reweighed. The water content of palm sugar can be calculated using the following equation:

$$\text{Water content} \, \% = \frac{m_0 - m_i}{m_i} \times 100\%$$

Where:

- Water content = water content (%)
- $m_0$ = initial sample mass (g)
- $m_i$ = final sample mass (g)

### 2.4.7. Yield

The yield was used to see the efficiency of the palm sugar tool by weighing the coconut sugar ingredients used to make palm sugar and weighing the final result of palm sugar. The yield value of palm sugar can be calculated using the following equation:
\[ \eta_p = \frac{m}{m_{in}} \times 100\% \]  \hspace{1cm} (26)

Where:

- \( \eta_p \) = product efficiency (%)
- \( m \) = total mass of the product produced (kg)
- \( m_{in} \) = total mass of the incoming material (kg)

2.5. Data analysis

The data analyzing process was done using IBM SPSS version 25.0 software and Microsoft Excel software. Data analysis is used to determine whether the type of tools used affect the rate of change in temperature, brix and density. The data analysis was done using a statistical test, by testing the homogeneity of the data to find out whether the analyzed data had similarities. When the significance value limit of the data homogeneity was > 0.05, then we could conclude that the data was homogeneous. Then the data was analyzed using one-way or two-way ANOVA, and for the posthoc test we used the Duncan multiple range test. When the significance value was < 0.05 then the treatment of the tool could affect the coefficient of temperature change, brix, density, fineness modulus, average particle diameter, color, moisture content and yield.

3. RESULT AND DISCUSSION

Palm sugar is a diversification of brown sugar products in the form of small granules (granulations) or crystalline powder with diameters ranging from 0.8 to 1.2 mm [3]. The result of the calculation of the analysis palm sugar can be seen in Table 4.

Table 4. The result of the analysis of the production palm sugar

<table>
<thead>
<tr>
<th>Process</th>
<th>Value</th>
<th>Variable</th>
<th>GSPE</th>
<th>GSKP</th>
</tr>
</thead>
<tbody>
<tr>
<td>Evaporation</td>
<td>( k_T ) (°C/minute)</td>
<td>0.007±2\times10^{-3}</td>
<td>0.004±2\times10^{-3}</td>
<td></td>
</tr>
<tr>
<td></td>
<td>( \rho ) (kg/m³/min)</td>
<td>2.08±10^{-4}</td>
<td>2.73±10^{-4}</td>
<td></td>
</tr>
<tr>
<td></td>
<td>nA</td>
<td>1.84±0.04</td>
<td>1.85±0.02</td>
<td></td>
</tr>
<tr>
<td>Crystallization</td>
<td>( k_T ) (°C/minute)</td>
<td>0.109±0.01</td>
<td>0.116±0.01</td>
<td></td>
</tr>
<tr>
<td></td>
<td>( \rho ) (kg/m³/min)</td>
<td>1.70±10^{-4}</td>
<td>1.67±10^{-4}</td>
<td></td>
</tr>
<tr>
<td></td>
<td>nA</td>
<td>1.86±0.10</td>
<td>1.87±0.03</td>
<td></td>
</tr>
<tr>
<td></td>
<td>( \delta ) (%/minute)</td>
<td>1.18±10^{-4}</td>
<td>1.07±10^{-4}</td>
<td></td>
</tr>
<tr>
<td></td>
<td>nA</td>
<td>1.92±0.03</td>
<td>1.94±0.16</td>
<td></td>
</tr>
<tr>
<td></td>
<td>FM</td>
<td>4.53±0.67</td>
<td>3.95±0.36</td>
<td></td>
</tr>
<tr>
<td></td>
<td>D (inch)</td>
<td>0.101±0.04</td>
<td>0.065±0.01</td>
<td></td>
</tr>
<tr>
<td>Color</td>
<td>L*</td>
<td>58.7±1.1</td>
<td>56.8±2.9</td>
<td></td>
</tr>
<tr>
<td></td>
<td>a*</td>
<td>18.88±0.6</td>
<td>16.35±1.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>b*</td>
<td>38.07±2.5</td>
<td>35.07±1.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Water content (%)</td>
<td>1.79±0.3</td>
<td>1.38±0.6</td>
<td></td>
</tr>
</tbody>
</table>

Mean values ± standard deviation from two-way analyzed. Superscripts in the column indicate not significant difference (P > 0.05) amount samples.

3.1. Temperature

The obtained value of the constant decrease in temperature change (\( k_T \)) affected the heat released by the higher value material (\( k_T \)) the temperature of the material decreased so fast so that the equilibrium between the temperature of the material and the ambient temperature was quickly achieved. The constant value of decreasing temperature in palm sugar was 0.18 °C/minute [4].

3.2. Brix

Brix is the amount of dissolved solids in a solution or a material. During the evaporation and crystallization process there was an increase in the brix content due to heating so that the temperature of the material increased. Then the water in the material evaporated and the solution became more concentrated and the solid increased.
3.3. Density
The rate of increase in density was directly proportional to the rate of increase in brix content. The density value during the evaporation process would increase continuously until it started to be constant. In the crystallization process there was a decrease in the density value, this happened because the initially viscous solution turned into sugar crystals. The higher the constant value of the change in density reduction \( k_p \) the faster the temperature decreased and the faster the crystal formation occurred.

3.4. Particle size
The fineness modulus is a number that represents the average particle size of the product. The smaller the fineness modulus, the finer the granules are, which causes the particle diameter to become smaller. The FM value was influenced by the speed and the continuity of the stirring method. Processing the palm sugar with the manual stirring method made the crystals of the produced palm sugar different from one another and typically larger in size because the speed and strength of stirring of each individual was different, while processing the palm sugar with the rotary crystallizer was more stable so that the sugar crystals produced were same. The value of particle diameter obtained based on the research met the standards of palm sugar exporters to several countries. The sugar grain size was 12-18 mesh, which was in the range of 0.074-0.0315 inches [5].

3.5. Color
The color index value showed a relatively high L* value, which was > 50 which means bright, with an a* index of between 14.46-19.29 which means red [6] and a b* index of 30-45 which means brownish yellow [7]. The results of the research on the color of palm sugar produced were brownish yellow in accordance with Indonesian National Standard 01.3743:1995. The yellow or brown color produced in palm sugar was due to the Maillard reaction and caramelization during the palm sugar processing.

3.6. Water/Moisture content
The water content of sugar is very influential on the storability of palm sugar, so monitoring the water content is very important. The water content in the material played a role in chemical reactions, enzymatic changes, and the growth of microorganisms. High water content caused the texture of the product to become moist, triggering the occurrence of sugar clumping (clumping) so that it could reduce the physical quality of the product [8]. The yield of palm sugar from all treatments on the water content was less than 3% which means it was in accordance with Indonesian National Standard 01.3743:1995.

3.7. Yield
The result of the yield percentage value is 78% so that the efficiency of the pan evaporator and rotary crystallizer.

4. CONCLUSION
Based on the results of the research it can be concluded that:
1. Constant changes in temperature, brix, and density in the evaporation process, respectively
0.004-0.007 °C/minute; 1.07 \times 10^{-3} \pm 0.3 \times 10^{-4} - 1.18 \times 10^{-3} \pm 1 \times 10^{-4} \% / \text{minute}; 2.08 \times 10^{-4} \pm 4 \times 10^{-5} - 2.73 \times 10^{-4} \pm 4 \times 10^{-5} \text{ (kg/m}^3) / \text{minute} \) and the nA values ranged from 77–1.94.

2. The crystallization process the temperature and density constants ranged from 0.109±0.01-0.116±0.01°C/minute; 1.67 \times 10^{-3} \pm 0.5 \times 10^{-4} - 1.7 \times 10^{-3} \pm 4 \times 10^{-4} \text{ (kg/m}^3) / \text{minute}.

3. Characteristics of palm sugar showed water content 1.38±0.6-1.79±0.3 \%, yield 78.42±3.6-79.13±3.4 \%, fineness modulus 3.95±0.36-4.53±0.67, particle diameter 0.065±0.01-0.101±0.04 inches, and the color the color brownish yellow.

4. Pan evaporator and rotary crystallizer can be used to improve or increase the quantity and quality of granulated sugar produced to be more hygienic, efficient, and effective, especially from the palm sugar processing industry which still applies conventional methods.

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REFERENCES


