Kinetics of the Coconut Sap Physical Properties During Palm Sugar Processing Using Pan Evaporator and Vertical Type Double Jacket Stirred Crystallizer

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ABSTRACT
Palm sugar is made from the evaporation and crystallization of palm sap turned into granules/powder. This research aims to analyze the physical kinetics of the coconut sap throughout the cooking process of palm sugar using pan evaporator and vertical type double jacket stirred crystallizer. Fresh coconut sap with a variety of capacities 5 litres, 10 litres, and 15 litres were cooked using pan evaporator until the sap saturated to thicken, following the process, the sap was crystallized using a vertical type double jacket stirred crystallizer. The crystallization process used a vertical type double jacket crystallizer until the palm sugar was formed. During the evaporation and crystallization process, the temperature, brix, and density were measured. The characterization of the product is conducted by water content, yield, particle size, and colour. Data on changes in physical properties were analyzed using avrami kinetics and 1-orde kinetics equations. The kinetics constants data were analyzed for their influence on the results of characterization. The results of the research on the evaporation process show that the temperature constant of $9.7x10^{-2}$ - $1.83x10^{-1}$°C/minute; density constants of $9.3x10^{-5}$-$1.1x10^{-4}$ kg/m^{3}minute; the brix constant of $1.5x10^{-6}$ - $9.8x10^{-5}$%/minute. The process of crystallization shows that temperature constants of 2.0 - $3.7x10^{-2}$°C/minute; density constant of 2 - $6x10^{-3}$ kg/m^{3}minute. Avrami kinetics and 1-orde can be used to predict the changes in physical properties during the process. The result of the physical characteristics of palm sugar shows that the water content appears 1.55-2.43%; the yield appears 13.57-24.37%; the fineness modulus appears 5.26-6.55 μm; the particle size appears 1.21-1.39 mm. In generals, the colour of palm sugar is brownish-yellow. The variations of capacity do not affect the physical quality of palm sugar.

Keywords: *crystallization, evaporation, modelling, palm sugar, physical quality*

1. INTRODUCTION

Sugar is one of the energy resources for the body, which leads the organs to work normally. Sugar has become the main trading commodity in Indonesia. Based on data from the Central Statistics Agency, the level of sugar consumption in Indonesia in 2017-2019 was 5.1 million tons [1]. National sugar production in 2021 is predicted to increase 5% from the previous year to 2.24 million tons. The Covid-19 pandemic has affected the level of low consumption of sugar. Even so, Indonesia continues to import sugar. It is due to the increasing demand but not accompanied by an increase in domestic production capacity [2].

Diversification of the variety of sugar products to reduce imports can be done by looking for some alternative raw materials other than sugar cane. One of which is sugar from the palm. The basic ingredients of sugar from palms include coconut sap, palm sap, siwalan sap, maple sap, and nipa sap [3]. The palm sap can be processed into liquid sugar, molded sugar, and palm sugar. Palm sugar is the result of processing from fresh sap (coconut/palm), which is heated to make thick sap, then stirred and gone through the process of cooling to form crystal granules.

Palm sugar has gained a lot of people's interest because it embodies many advantages, such as having a crystal or powder form so that it is easy to carry due to its safe packaging. Not only that, but it also has durable storage because the amount of water is lower compared to molded sugar. However, contrary to its advantages, palm sugar has some weaknesses, which is the process is relatively long and not easy. It is why its price in the market tends to be higher compared to molded sugar.

Currently, the processing of palm sugar is done conventionally with a wood-fired stove, using an iron pan which is not safe for food processing. The conventional processing of palm sugar has weaknesses, one of which is the use of a manual stirrer during the crystallization process. The manual mixing process requires more energy for mass production.

2. MATERIAL AND METHODS

This research is conducted on July 2020 – May 2021 at the Food and Postharvest Engineering Laboratory, Department of Agricultural and Biosystem Engineering, Faculty of Agricultural Technology, Gadjah Mada University.

2.1 Materials

The tools used for the palm sugar processing consist of the main tools and supporting tools for the data collection. The main tools consist of a pan evaporator and a vertical stirred double jacket crystallizer. Meanwhile, the supporting tools for the data collection consist of a thermometer, pH meter, refractometer, scales, measuring cup, and so on. The materials used in this study are fresh coconut sap. Fresh sap is taken from the sap vine farmers in Wonosari Hamlet, Jerukagung Village, Salam, Magelang, Central Java. The sap used for this research is the sap that has been cooked for 30 minutes to anticipate the decrease of acidity levels after the draining. Various materials used are several volumes of fresh sap of 5 liters, 10 liters, and 15 liters.

2.2 Palm Sugar Processing

The main process of palm sugar production is divided into 2 stages: the evaporation process and the crystallization process. Besides that, to dry granulated sugar from wet granulated sugar, there are two additional processes: drying and sifting.

2.2.1 Evaporation Process

The evaporation process is conducted by using a pan evaporator. Before the cooking process begins, make sure all of the measuring devices are prepared. The purpose of this evaporation process is to concentrate the juice. So, it changes from fresh juice to thick juice within the span of the cooking time with some adjustment of the initial capacity. This process starts with the filtering process to separate the impure substances and measure the pH value of the sap. A good sap has a pH value of 5-7. If the acidity rate is below 5, it can be predicted that the crystallization will fail. Based on Susi [4], if the juice has a pH of less than 6 or even tends to be acidic. Then, the sucrose within has been hydrolyzed into glucose and fructose. So, the crystallization process will not be good.

During the process of cooking and evaporation, the temperature parameters are measured every 3 minutes at the degree of body temperature. It includes the top of the material, the pan, the side of the pan, the combustion chamber, and the chimney. The Brix and density measurements during the evaporation process are measured every 5 minutes in the beginning until the Brix is 50% (towards thick). The Brix and its density are expected to be 50-60% formed into Brix crystal cores (granular/powder form), and if it increases quickly, the measurements are adjusted to every 3 minutes. When the sap is thick, the heat should be lowered down to make it not burn. The juice that is ready to be crystallized is the one with a supersaturated condition. It is indicated by the hairs or elongated threads created through the sap when removed using a wooden stirrer.

2.2.2 Crystallization Process

The crystallization process aims to form palm sugar particles from thick sap to become granular granules of palm sugar. The first stage of the crystallization process is to fill water into the crystallizer cylinder through a chimney of 10 L of water vapor. This stage can also be carried out at the beginning of the tools and materials preparation before the evaporation process begins. The water is added later as a heating source for the material during the further thickening process. The water that is added is then heated using a gas stove until the water boils. It is indicated by the hot steam that comes out of the hole on the edge of the pan. The material that is ready for the crystallization process is then put into the crystallizer pan.

During the crystallization process, the data of the temperature, Brix, and density are taken every 3 minutes. The crystallization process requires a rapid decrease in temperature. If the sap has turned to be thick, the stove is turned off, and the water drain valve is opened, so the heat of the pan will be reduced. To make the sap crystallized, the process of decreasing the temperature needs to be maintained, in which the stirring process is carried out at a speed of 45 RPM. The stirring rotation is set not too fast or too slow, so the material particles can collide with each other to form palm sugar crystals. The crystallization process lasts for 30-60 minutes until all the materials are crystalline and the material temperature decreases to 40-50.

2.2.3 Drying Process

When left alone, wet palm sugar products will damage both the physical structure and chemical structure, along with the increase of the water density due to collisions between particles that are not yet dry. Therefore, after the crystallization process is complete, the drying process is carried out using a cabinet dryer at a temperature of 60°C for 3-4 hours. The purpose of the drying process is to reduce the water content of the product to a maximum of 3%. Dry products will facilitate the sifting process.

2.2.4 Sifting Process

The sifting process of palm sugar is carried out after the sugar product is dried. It makes the product that has a high-water density does not stick to the sieve holes. The sieving process is carried out using a Tyler sieve with mesh sizes of 8, 14, 30, 40, 50, and 100. The sifting process of granulated sugar products is conducted to equalize the particle size and determine the fineness modulus (FM) value or the degree of the product fineness.2.3 Analytical Methods

The data are analyzed using Avrami kinetics, first-order kinetics, and statistical analysis.

2.3.1 Determination of the Constant Changes of the Temperature, Brix, and Density during the Evaporation Process

a. Temperature Change Constant

In the evaporation process, the value of the temperature change is determined using the Avrami equation. The data of the temperature change (1) is measured by changing the values of k and nA to obtain the predicted temperature value.

$$T_t = \left(-1 + \left(exp\left(-k.t^{nA}\right)\right)\right).\left(T_e - T_0\right) + T_0 \quad (1)$$

Where:
T_e = Final Temperature
T_0 = Initial Temperature

b. Brix Change Constant

Brix is a parameter that states the number of dissolved solids in a solution. The amount of dissolved solids is usually expressed in percent (%). The Brix measuring device is called a refractometer. Brix is not a determinant of the pure sucrose density in a solution, but the total amount of solids in a solution contain sugar [5]. During the process of producing palm sugar, Brix data collection is taken every 5 minutes in the beginning until the Brix is 50% (sap begins to get thick); then every 3 minutes, if the sap has already grown thick until it becomes crystal sugar. It is indicated by the Brix scale refractometer of 0-85%. Furthermore, once the Brix is not readable by the measuring instrument, then the dilution process can be conducted.

$$B_\theta = \frac{m_{t+g} - m_g}{m_{0+g} - m_g}.B_p \quad (2)$$

B_θ is the actual amount of brix, while B_p is the brix reads on the refractometer after dilution. m_{t+g} is the mass after dilution, m_{0+g} is the mass before dilution, and m_g is the mass of the measuring cup.

Next, the brix data is analyzed using the Avrami Model (3-5) to determine the constants during the evaporation and crystallization process.

$$\frac{B_\theta - B_e}{B_0 - B_e} \approx \frac{B_t - B_\theta}{B_e - B_0} \quad (3)$$

$$B_t - B_0 = ((1 - (exp\,(-k.t^{nA}\,)\,).(B_e - B_0) \quad (4)$$

$$B_t = (1 - (exp\,(-k.t^{nA}\,)\,)).(B_e - B_0) + B_0 \quad (5)$$

c. Density Change Constant

Density is the specific gravity (ρ) by measuring the mass (m) and the volume of the material (v) (6). The density measurements are conducted every 5 minutes until the brix reaches 50% and a span of 3 minutes after the brix has gained the degree of 50%. The measurement of density uses a 10 ml measuring cup, in which the material is added to a volume of 10 ml, and then the mass is weighed.

$$\rho = \frac{m}{v} \quad (6)$$

The density data calculation is then modeled using the Avrami Model (7-10) to determine the predicted density value during the evaporation process.

$$\frac{\rho_\theta - \rho_e}{\rho_0 - \rho_e} \approx \frac{\rho_t - \rho_0}{\rho_e - \rho_0} \quad (7)$$

$$\frac{\rho_t - \rho_0}{\rho_e - \rho_0} = (k.t^{nA}\,)) - 1) \quad (8)$$

$$\rho_t - \rho_0 = (k.t^{nA}\,)) - 1)\ .(\rho_e - \rho_0) \quad (9)$$

$$\rho_t = (k.t^{nA}\,)) - 1)\ .(\rho_e - \rho_0) + \rho_0 \quad (10)$$

2.3.2 Determination of the Constant Changes of the Temperature, Brix, and Density during the Crystallization Process

a. Temperature Change Constant

During crystallization, the predicted temperature is calculated using first-order kinetics (11-15). This is because the process is directly proportional to time.

$$\frac{dx}{dt} = \pm k.x \quad (11)$$

$$\frac{dx}{x} = \pm k.dt \quad (12)$$

$$\int_0^t \frac{dx}{x} = \int_0^t \pm k.dt \quad (13)$$

$$ln\,x_t - In\,x_0 = \pm k.t \quad (14)$$

$$In\,(\tfrac{x_t}{x_0}) = \pm k.t \quad (15)$$

b. Brix Change Constant

The data of the brix change in the crystallization process is measured using (5). Final brix is denoted by B_e (%), whereas initial brix is denoted by B_0 (%).

c. Density Change Constant

The data of the density change in the crystallization process is measured using (16).

$$\rho_t = \rho_0 - (k.t^{nA})) - 1) .(\rho_0 - \rho_e) \quad (16)$$

2.3.3 Statistics Analysis

The data obtained are statistically analyzed using SPSS 25 software. The analysis used is Two-Way ANOVA with the Duncan Multiple Range model. Previously, the homogeneity of the data is tested using the Levene test. Parameters measured include temperature, Brix, density, yield, moisture content, size distribution (FM), and color. After that, the first-order kinetics is used for the temperature data analysis. Meanwhile, Avrami kinetics is used for the analysis of Brix and density values. Both kinetic analyzes are calculated using Excel 2013 software.

2.3.4 Distribution of the Particle Sizes

Fineness modulus indicates equality in grinding the resultant product. Fineness Modulus (FM) is defined as the sum of the weight fractions retained over each sieve divided by 100 (17).

$$FM = \frac{\sum iXi}{100} \quad (17)$$

Furthermore, the average diameter of the particles in mm can be measured using (18)[6].

$$D = 10^{(\frac{\sum m_i logDi}{\sum m_i})} \quad (18)$$

2.3.4 Yield

Yield is the ratio of the mass of the product produced to the mass of the material used. The unit to express the yield value is percent (%). Many factors affect the yield value, including the quality of raw materials, the production process procedures, and the quality of machinery and equipment [7]. The higher the yield value, the more products are produced. The value of the yield can be measured with (19).

$$Yield\ (\%) = \frac{output\ mass}{input\ mass} x100\% \quad (19)$$

The output mass is the mass of the processed granulated sugar product, while the input mass is the mass of the fresh sap material that is ready to be processed.

2.3.5 Water/Moisture Content

Moisture content is the water content within the material (20). Based on SNI, palm sugar has a maximum water content of 3%. To achieve this moisture content, a drying process is needed.

$$Moisture\ Content\ (\%) = \frac{w - w_1}{w - w_2}\ x\ 100\% \quad (20)$$

w is the sum of the mass of the cup and the mass of the initial material before the drying process (grams), w_1 is the sum of the mass of the cup and the mass of the material after drying (grams), while w_2 is the mass of the empty cup (grams).

2.3.6 Color

Color measurement can be done by a digital refractometer. One of the measurement methods is the CIELAB method which is denoted by 3 signs, namely L*, a*, and b*. The CIELAB color chart is shown in Figure 1. The L* value of 0 indicates black, and 100 is white. The notation a* indicates a mixed red-green color if the value is +a* for red and -a* for green. The notation b* indicates a mixture of blue-yellow colors, if +b* for yellow and -b* for blue [8].

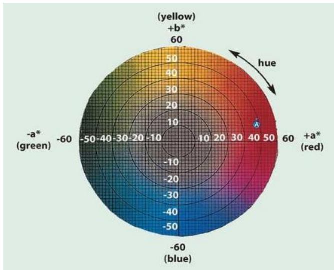

Figure 1. CIELAB Diagram

3. RESULTS AND DISCUSSIONS

3.1 Temperature Changes in Evaporation

At the beginning of the evaporation process, the temperature of the material increases significantly. It is because of the sensible heat received to increase the temperature of the material. The material experiences an increase in temperature until it reaches its boiling point so that the temperature becomes constant and the material becomes viscous due to the sub heat. Aksar [9] explained that sensible heat is heat added to an object that is heated so that the object's temperature will rise. Meanwhile, sub heat is the heat absorbed by the material during the process at a constant temperature.

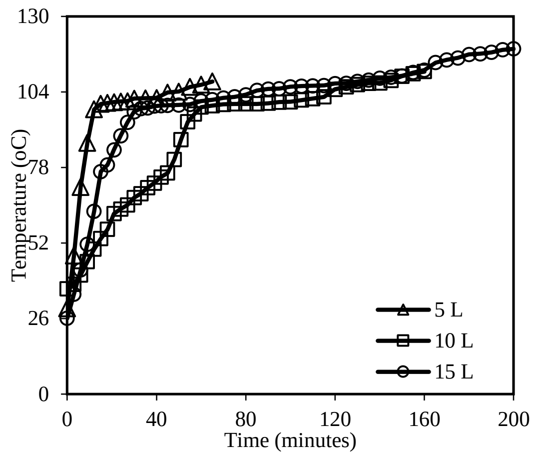

Figure 2. Temperature changes during evaporation

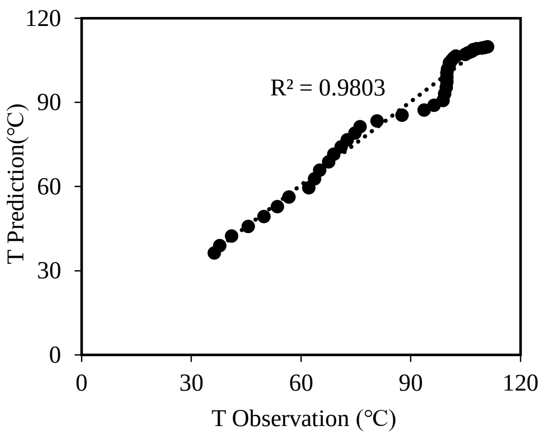

Figure 3. Evaporation process of $T_{observation}$ and Tprediction Validation

Table 1. Constant changes of temperature (kT) during Evaporation Process

Treatment	k_T (°C/minutes)
GSM5	0.183±0.092[b]
GSM10	0.097±0.078[a]
GSM15	0.167±0.042[a]

The mean ± standard deviation of the 3 replicates superscript analysis is the same, not significantly different (Significant < 0.05).

In this study, the capacity or mass of the cooked material influences the rate of temperature changes. If the capacity is small, the rate of temperature changes will be faster. It is because the water contained in the material evaporates quickly. Rosmindari [10] stated that the value of k_T can be influenced by mass because the more material is processed, the slower heat transfer process will be so that the k_T also will be smaller.

3.2 Temperature Changes on Crystallization

The crystallization process takes place after a further heating process of saturated juice that becomes supersaturated juice in the crystallizer. Crystal growth can happen along with a decrease in temperature. Therefore, the heating process on the crystallizer is stopped, and the water heating source is discharged through the faucet. The stirring speed also influences the crystallization process. That is why the speed of the rotary stirrer is set neither too slow nor too fast. The speed used during the constant crystallization stirring process is 45 RPM.

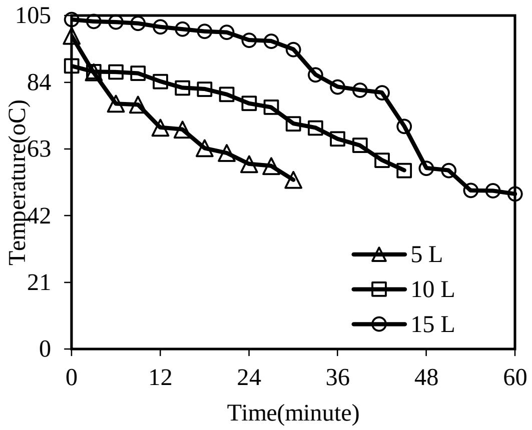

Figure 4. Temperature changes during crystallization

The data obtained have a decreasing trend because crystal growth can form along with the rate of decrease in the temperature of the material. The heat from the material is released into the environment. It is to reach an equilibrium between the temperature of the material and the environment. The heat transfer that occurs in the crystallization process is by convection. However, the temperature of the steam and heating water are lowered slowly. That is why the temperature of the material significantly does not drop, which affects the resulting crystals.

3.3 Brix Change during Processing

Brix is measured every 5 minutes in the evaporation process and every 3 minutes in the crystallization process. The first material, called fresh sap, contains a lot of water content, so the value of the Brix content is still low. Along with the heating processes taking place, the material will thicken and evaporate to form a solid. Therefore, the Brix value will increase until it reaches a constant, which means the sap material is already in a saturated condition. Thick sap's color is brown due to the caramelization reaction and the Maillard reaction [11].

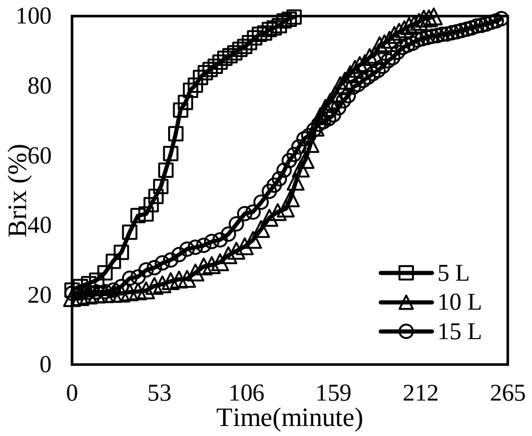

Figure 5. Brix changes during process

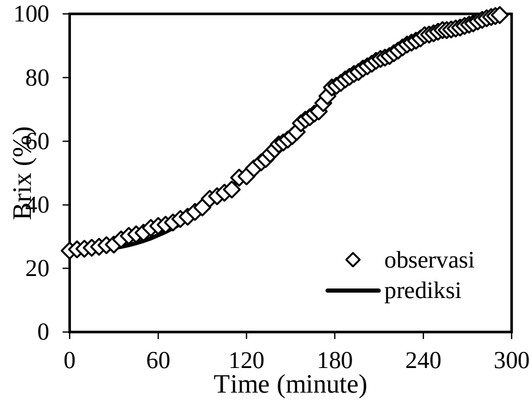

Figure 6. Relationship between Observation Brix and Prediction Brix

Table 2. Constant changes of brix (kB) during cooking process

Treatment	k_B (%/minutes)
GSM5	$9.83 \times 10^{-5} \pm 0.319^{a.b}$
GSM10	$0.27 \times 10^{-5} \pm 0.342^{a}$
GSM15	$0.15 \times 10^{-5} \pm 0.348^{b}$

The mean ± standard deviation of the 3 replicates superscript is the same, not significantly different (Significant < 0.05)

Two-way ANOVA statistical analysis data shows that the variation of capacity treatment has no significant effect on the rate coefficient value of Brix changes (kB). It can be seen that the subsets obtained are almost the same.

3.4 Density Changes on Evaporation

Changes in the temperature of the material due to a heat source cause the water content to evaporate to become saturated juice (thick juice), and the concentration of solid increases. The temperature changes that occur affect the density changes. The material loses some water, but the mass of the material increases with the cooking process. Kusuma [12] stated that the reduced mass of material (water) also decreases. Yet, the volume shrinkage occurs more so that the density of sap during cooking increases.

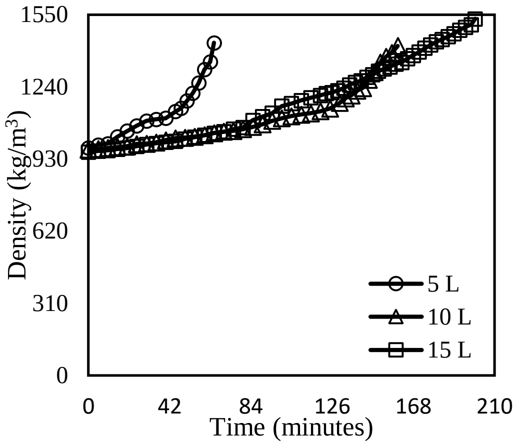

Figure 7. Density changes during evaporation

Table 3. Constant changes of density (kρ) during evaporation process

Treatment	$k\rho$ (kg/m^3minutes)
GSM5	$11.2 \times 10^{-5} \pm 8.0 \times 10^{-6b}$
GSM10	$10.1 \times 10^{-5} \pm 0.0^{b}$
GSM15	$9.3 \times 10^{-5} \pm 6.2 \times 10^{-6a}$

The constant density with 5 liters and 10 liters cooking shows the same subset. It means that they are not significantly different. However, the 15 liters capacity represents a different subset, as 15 liters cooking takes longer. In the cooking process of palm sugar with more capacity, the rate of density change to reach equilibrium (supersaturated juice) will be longer. It is because coconut sap contains high water fairly content, which is around 88.4% [13].

3.5 Density Changes on Crystallization

The crystallization process occurs along with the growth of palm sugar crystals. One of the factors for crystal growth is density. Density is the ratio of mass per unit volume. In the crystallization process, the density data is taken every 3 minutes. It is because the intermolecular separation happens as the molecules are colliding in the stirring process. Therefore, the process of decreasing the temperature of the material is also faster. And this process affects the density value.

Table 4. shows the same subset of each treatment, which means there is no significant difference. The density value which taken as physical parameter of the material during the crystallization process will decrease along with the separation of solid particles, which will form palm sugar crystals.

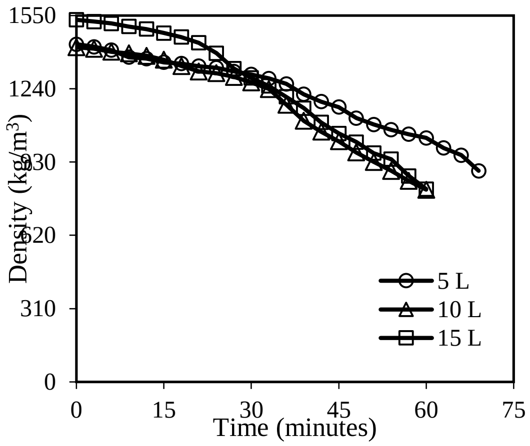

Figure 8. Density changes during crystallization

Table 4. Constant changes of density (kρ) during crystallization process

Treatment	$k\rho$ (kg/m^{3}minutes)
GSM5	0.006 ± 0.185[a]
GSM10	0.002 ± 0.080[a]
GSM15	0.002 ± 0.191[a]

3.6 The Physical Characteristics of Palm Sugar

Product durability is very much important, one of which is the water contained in the product. It affects the process of food spoilage. The water content of palm sugar produced ranges from 1.48-2.61%, as shown in Table 5, including good quality, because the water content in palm sugar is still in a recommended range by the Indonesian National Standard (1995), which is 3 % as the maximum water content of palm sugar.

Table 5. Water content value of palm sugar

Treatment	Test	Water Content (%)
GSM5	1	2.26
	2	2.61
	3	2.43
Average		2.43
GSM10	1	1.79
	2	2.02
	3	1.95
Average		1.92
GSM15	1	1.51
	2	2.45
	3	1.64
Average		1.87

Furthermore, the yield of palm sugar produced influences variations in capacity. The average of palm sugar obtained is 17.8%. According to Zuliana et al [11], the yield which is not high means that the palm sugar is quite difficult to crystallize, so that a lot of coconut sugar is left in the pan. The higher reduction sugar causes the more difficulty of crystallizing palm sugar. The higher the reduction sugar in palm sugar products, the higher the water content causing the lower yield.

One condition of palm sugar quality determined by Indonesian National Standard (SNI) is the color of the sugar. National Standardization Agency of Indonesia has set the standard of the sugar color, which is brownish-yellow to brown.

3.7 Application of Constant Changes in Physical Properties in Process Design of Palm Sugar

Palm sugar processing design can be done by applying the constants obtained during the processing, as shown in Table 6.

Table 6. Constant of the physical characteristic of palm sugar processing

Treatment	Evaporation			Cryztallization		Percentage passing mesh 14-40 (%)
	kT (°C/min)	kB (°C/min)	kρ (kg/m^{3}min)	kT (°C/min)	kρ (kg/m^{3}min)	
GSM5	1.83×10^{-1}	9.83×10^{-5}	1.12×10^{-4}	2×10^{-2}	6×10^{-3}	44.76
GSM10	9.7×10^{-2}	2.7×10^{-6}	1.01×10^{-4}	3.7×10^{-2}	2×10^{-3}	47.24
GSM15	1.67×10^{-1}	1.5×10^{-6}	9.3×10^{-5}	2.8×10^{-2}	2×10^{-3}	39.76

Based on the product value that passes the 14-40 mesh, the sugar products produced are under export standards, namely 12-18 mesh with the particle sizes between 0.187 – 0.99 mm[2].

4. CONCLUSION

The results of this research can be concluded that:

1. The constant of temperature changes on the evaporation process is 9.7×10^{-2} - 1.83×10^{-1} °C/minute; the constant of density changes is 9.3×10^{-5} - 1.12×10^{-4} kg/m^{3}minute; the constant of brix changes during the process of palm sugar production is 1.5×10^{-6} - 9.83×10^{-5} % /minute.
2. The constant of temperature changes on the crystallization process is 2.0×10^{-2} - 3.7×10^{-2} °C/minute; the constant of density changes is 2.6×10^{-3} - 6×10^{-3} kg/m^{3}minute.
3. The water content values of palm sugar is 1.55 - 2.43%; the yield value is 13.57 - 24.37%; the fineness modulus value is 5.26 - 6.55 μm; the particles size distribution is 1.12 - 1.39 mm; the color of the palm sugar is brownish-yellow.
4. The kinetics constant application of the physical characteristic during the evaporation

and crystallization process can be done in producing the palm sugar.

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