

Effect of Operating Temperature on Yield of Liquid Products in the Pyrolysis Process of Used Tires into Fuel Oil

Muhammad Irsan. B^{a,1,*}, Hasbi Assiddiq S.^{b,2}

^a Department of Electrical Engineering, Politeknik Kotabaru, Stagen, Kotabaru 72114, Indonesia

^b Department of Mechanical Engineering, Politeknik Kotabaru, Stagen, Kotabaru 72114, Indonesia

¹ irsancanoa@gmail.com *; ² hasbiassiddiq999@gmail.com

* corresponding author

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ABSTRACT

Used tires are one of the wastes generated from both two-wheeled and four-wheeled vehicles. Indonesia is one of the largest producers of used tire waste because vehicle users are increasing. The utilization of used tire waste is still very small, so a method is needed to convert used tire waste into valuable materials. One method that can be used to transform used tire waste is the pyrolysis method which can convert used tire waste into fuel. The pyrolysis process of waste tires uses raw materials from used tires that have been cut into pieces with a size of 10 × 10 cm. The waste tires used are two-wheeled (motorcycle) tires. This process uses temperatures in the range of 400-600 °C. The temperature range is to find the most optimal operating temperature that can be used to convert used tire waste into liquid products without using a catalyst. Waste tires that have been cut to the desired size are put into a batch reactor of 2 kg and heated to a predetermined temperature of 400, 450, 500, 550, and 600 °C for 1 hour. In this study, two types of products were obtained: liquid and solid. The liquid product results from the gas cooling process produced by using a condenser. Under these operating conditions, the yields of liquid products at temperatures of 400, 450, 500, 550, and 600 °C were 39.32; 39.02; 42.58; 42.11; and 41.84 %wt, respectively. The process is carried out for 1 hour in the reactor by maintaining a predetermined operating temperature. The solid product obtained is residual pyrolysis charcoal which cannot be decomposed with the yield obtained at temperatures of 400, 450, 500, 550, and 600 °C, respectively, 40.73; 41.54; 38.82; 37.68, and 35.91 %wt.

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1. Introduction

Since the presence of the rubber industry, technology and chemists have developed materials and products that have high resistance, heat resistance, and resistance to aggressive environments; consequently, they must have high performance and long life. Rubber waste (usually whole or torn tires) can be reduced to gas, oil, charcoal, metal, and inorganic fractions. Further processing can then be used as an energy source or as additives and starting materials for other products [1]. Used tires are waste mainly produced in developing countries, especially in Indonesia. This is due to the increasing number of private vehicle users yearly. Based on data from the Central Statistics Agency in 2019, the use of rubber, especially for the most widely used vehicles, namely motorbikes and cars, as many as 811,136 pieces per unit, with details of service, namely 34,086 motorcycle tires per unit, 159,018 motorcycle tires per unit, and outer tires for cars as much as 618,032 pieces per unit [2]. From this data, it can be estimated that the use of private vehicles can cause a buildup of used tire waste, so there is a need for a process to recycle used tire waste which is

Increasing from year to year. There has been a growing awareness of the potential environmental impact of the tire industry in the last two decades. As an efficient technique, life cycle assessment (LCA) is applied to measure the environmental impact of industrial tires through four life cycle stages, namely, production, transportation, use, and end of life [3]. One method that can be used to reduce waste tire waste is the pyrolysis process. The process that can be used to minimize used tire waste is to convert used tire waste into fuel as an alternative to solve problems regarding energy and the environment. Several methods can be used to decompose used tire waste into fuel, one of which is the pyrolysis process to convert used tire waste into oil.

Pyrolysis or thermolysis is an irreversible thermochemical process that is very important for complex solid materials or chemical compounds involving an increase in temperature in oxygen-free conditions. The pyrolysis process is highly dependent on operating temperature. During the pyrolysis process, the molecules are subjected to very high temperatures, which makes the molecules experience very high vibrations where the molecules are stretched and shaken so that the molecules can break down into smaller molecules [4]. In slow pyrolysis or conventional pyrolysis, the raw material is heated slowly (slow) at a heating rate (0.1 – 2 per second) at a low pyrolysis temperature (< 400 °C) for a long time. Slow pyrolysis process of biomass, biomass is devolatilized to produce tar and charcoal as the main products. The resulting gas consists mainly of methane and small amounts of hydrogen, propane, ethylene, CO, and CO₂ [4], [5]. High heating speed, high temperature, and short residence time produce high gas production. High heating speed, medium temperature, and temporary residence time result in higher liquid products [5]. In the pyrolysis of waste tires without a catalyst, the gas yield increases, and the charcoal yield decreases with increasing reaction temperature. The maximum yield is 54% at 500 °C; at 400 °C the charcoal yield is relatively high (32 %wt) and reduces until it reaches (18 %wt) at 600 °C [6]. Pyrolysis of waste tires is used to decompose waste tires at very high temperatures without oxygen [7], [8]. The liquid product produced in the pyrolysis process of used tires is a thick black liquid, which looks like a petroleum fraction [9].

A batch reactor was used in this study to obtain the desired optimal operating conditions for the production of fuel using waste tires. A batch reactor is used because it is under the main objective of the research, which is to determine optimal operating conditions in the production process of fuel oil using waste tires. Batch reactors are used for laboratory scale to find the optimal operating temperature variable. The desired product in this research is liquid, and the selected operating conditions are in the range of 400 – 600 °C. If the operating temperature used exceeds 400 – 600 °C, the resulting product is more dominant in gas; similarly, if the operating temperature is less than 400 – 600 °C, the resulting product is more dominant in solids. This study aims to find the optimal operating temperature to produce liquid products without a catalyst.

2. Research Methods

The research method used in this study is to utilize a batch reactor as a reactor used to convert waste tires into fuel oil. The pyrolysis process in the reactor will last 1 hour at a temperature range of 400-600 °C. The series of batch reactors used can be seen in Fig. 1, where the heater used is a fire heater (furnace) using a high-pressure burner fueled by LPG gas.

2.1. Tools and Materials

The tool used is a batch reactor with a maximum volume of 30 L and is equipped with a condenser to cool the resulting liquid product until it reaches room temperature (30 °C). The heating process in this reactor uses a fire burner (stove) with LPG gas as fuel. Materials used in the pyrolysis process of used motorcycle tires and LPG gas as fuel. Motorcycle tires (outer) are used because they are easier to obtain, and the waste treatment process is easier than car tires. Tires that are processed in the reactor are 2 kg in one pyrolysis process. The raw material for waste tires is put into the reactor tank, and then the burner is turned on as a heater used in this study. From Fig. 1, the reactor tank has a maximum capacity of 30 L. The process occurs in a reactor tank with a predetermined temperature of 400-600 °C. Waste tires that have been reduced in size according to the required size (10 × 10 cm) are put into the reactor tank, and then the reactor tank is tightly closed. The stove is then turned on and waited until it reaches the desired temperature. When the operating conditions

have been reached, the operating temperature is maintained for 1 hour to obtain the desired liquid product. The liquid produced in the reactor tank will exit through the pipe above and pass through the condenser so that the resulting liquid is condensed and then enters the liquid product reservoir.

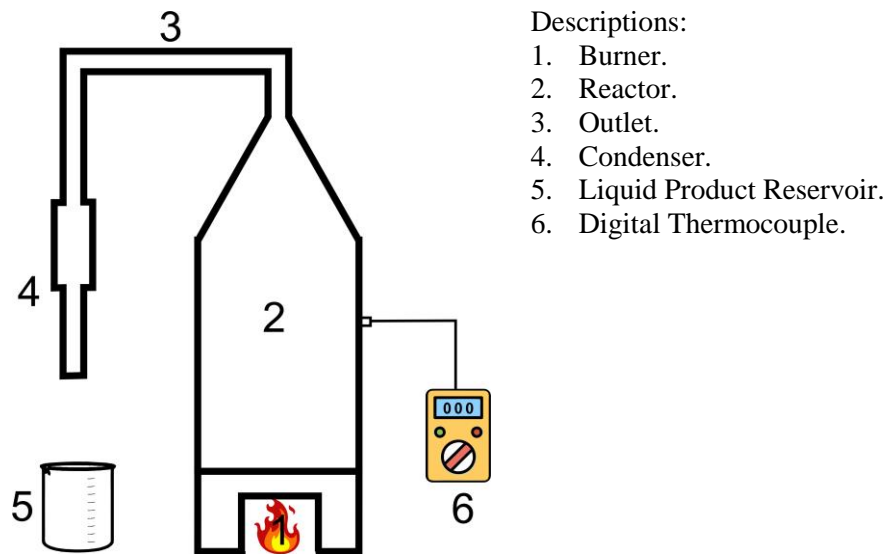


Fig. 1. Pyrolysis Reactor.

2.2. Raw Material Preparation

The raw material used is motorcycle tire waste which is used as the sole raw material in the pyrolysis process of used tire waste into fuel oil. Waste used tires are prepared in advance by reducing the size of the used tires to a smaller size, namely 10×10 cm. The tires that have been reduced in size are cleaned, and first of all, the dirt on the used tires. The process is continued by entering the cleaned raw materials into the reactor. Waste tire management aims to identify the most efficient approach to limiting the impact of such waste on the environment. Consumption reduction, reuse/recycling, and energy recovery are strategies used to solve the tire waste problem, as shown in Fig. 2 [10].

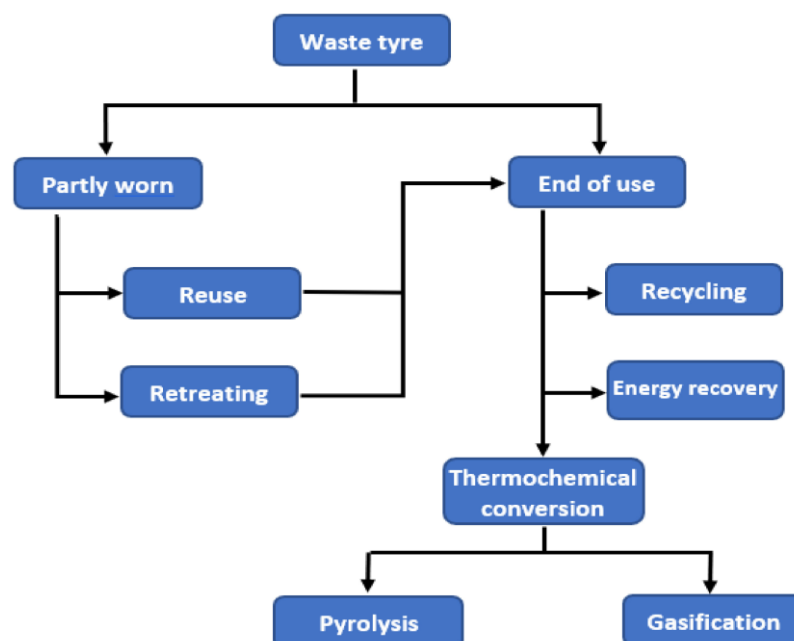


Fig. 2. Types of used tire management system [10].

2.3. Pyrolysis Process

Pyrolysis of used tire waste is profitable and depends on various factors, including product quality, production efficiency, and overall production costs. The composition of gas and oil from TPO (Tires Pyrolytic Oil) is highly dependent on the type of reactor used, where the heating rate and operating temperature determine the composition of oil and gas products [11]. Charcoal from the waste tire pyrolysis process generally contains carbon black and additives and has high thermal and stable mass. All charcoal has ash content in the 13-16 %wt, and oxygen (O) content has a content range of 13-20 %wt. The decrease in O content in charcoal with increasing temperature indicates that additive decomposition occurs at higher pyrolysis temperatures [12]. The average single-passenger vehicle tire has between 30,000 kJ/kg and 35,000 kJ/kg energy potential, much higher than coal and biomass of the same mass, with lower ash content [13].

Waste tires that have been cleaned are then put into a reactor prepared to convert them into fuel oil. The pyrolysis process consists of three (3) progressive stages: initiation, propagation, and termination. Initiation reactions break down large polymer molecules into free radicals. The propagation reaction then breaks down the free radicals and molecular species into smaller radicals and molecules. The termination process will recombine radicals into more stable molecules [14]. The pyrolysis process lasted 1 hour by maintaining the desired operating temperature at 400, 450, 500, 550, and 600 °C. In this pyrolysis process, the desired product is liquid, so it uses operating conditions of 400-600 °C. Waste tires in the reactor will be processed based on a predetermined temperature range for 1 hour in a batch reactor. The raw materials are processed in the reactor, and gas is produced. The resulting gas is then condensed through a condenser to obtain two types of gas products: gas that can be condensed into liquid (oil) and gas that cannot be condensed (syn-gas). The condensation process occurs using air at room conditions ($T = 30\text{ °C}$ and $P = 1\text{ atm}$). The process utilizes pipes made of stainless steel so that cooling with air at room temperature can occur so that a liquid product (oil) is obtained as a result of the condensation process of gas products. The operating conditions will be sought for optimal conditions that produce the highest yield without using a catalyst.

2.4. Pyrolysis Product

Determination of the yield of the resulting product using equation (1) for liquid products, equation (2) for solid products, and equation (3) for gas products. There are three types of products: solid, liquid, and gas. Solids are produced from the rest of the pyrolysis process of materials that do not decompose, so that solid waste is obtained in the form of charcoal. The liquid product is obtained from the gas produced in the pyrolysis process, which can be condensed into a liquid through a condenser. The product gas is produced from non-condensable gas through the condenser. The gas produced from the pyrolysis process is in the form of light hydrocarbon gas (C1-C4), with methane and hydrogen gas having the highest concentration and hydrogen gas having the highest heating value [7], [15]. The yield of the product (solid, liquid, and gas) can be determined using equations (1), (2), and (3) based on the product obtained [16].

$$\text{Liquid yield (\%wt)} = \frac{m_{\text{liquid}}}{m_{\text{raw material}}} \times 100\% \quad (1)$$

$$\text{Char yield (\%wt)} = \frac{m_{\text{char}}}{m_{\text{raw material}}} \times 100\% \quad (2)$$

$$\text{Gas yield (\%wt)} = 100\% - [\text{char yield (\%wt)} + \text{liquid yield (\%wt)}] \quad (3)$$

3. Result and Discussion

Waste tires that have gone through the pyrolysis process produce liquid, solid, and non-condensable gas (gas) products. The main product produced from the pyrolysis of waste tires is condensable gas that forms pyrolysis oil (pyrolysis oil or pyrolytic oil). Pyrolysis oil is a complex mixture of hydrocarbons containing high concentrations of aromatic compounds. The primary use of pyrolysis oil is an alternative fuel due to its favorable hydrocarbon composition [17]. Liquid products in the form of oil and solid products in the form of charcoal left over from pyrolysis that has been carried out on the waste tires. The product of this pyrolysis process can be seen in Fig. 1 and Fig. 2. The results of the pyrolysis process of motorcycle waste tires obtained three types of products: solid, liquid, and gas. Rubber is an example of an elastomeric type polymer, a polymer that can return to its original shape after being stretched or deformed.

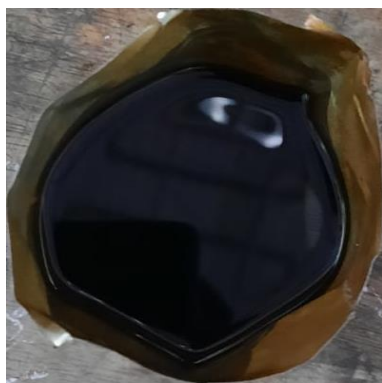


Fig. 3. Liquid Product.

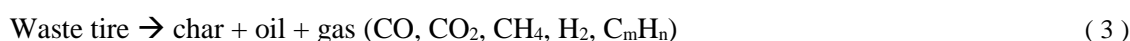


Fig. 4. Solid Product.

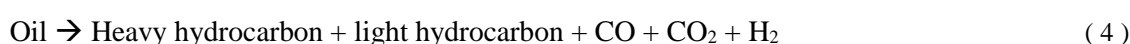
The research shows 3 product forms: solid, liquid, and gas (non-condensable gas). 2 products can be obtained from this pyrolysis process, as shown in Fig. 3 and Fig. 4. The liquid product can be seen in Fig. 3. The resulting oil is thick black with a fairly sharp aroma. The solid product can be seen in Fig. 4. The remaining waste tire waste is not completely decomposed, and some has been completely decomposed so that a solid product is obtained in the form of charcoal. The product gas (non-condensable gas) cannot be analyzed because the equipment is inadequate to capture the gas produced. The most desirable products are liquid products such as fuel oil. The pyrolysis process takes place at a temperature of 400-600 °C with a residence time in the reactor of 1 hour. The residence time in the reactor was determined by testing the reactor during the pyrolysis process. The results were obtained 1 hour after the pyrolysis process took place; the pyrolysis process did not produce oil, so the residence time in the reactor was used for 1 hour.

3.1. Pyrolysis Process

Pyrolysis is a thermochemical process that occurs at increasing temperatures and humid atmospheric conditions, where waste tires are converted into solid residues rich in carbon (charcoal), by releasing volatile substances (oil + gas), which can be described according to the reaction equation following [9]:



The pyrolysis oil will increase with increasing temperature until the maximum pyrolysis oil is obtained at a temperature of 500 °C, after which the oil produced decreases at temperatures above 500 °C. Temperatures above 500 °C will produce more gas (non-condensable gas) so that less oil is produced [9]:



A synthetic rubber that is usually used in the manufacture of tires is styrene-butadiene rubber (SBR), followed by polybutadiene rubber (BR) [18]. The composition of the rubber in the crushed waste tires can be seen in table 1 [19]:

Table 1. Composition of rubber in waste tires [19]

Element analysis	
C (wt. %)	84.3
H (wt. %)	7.7
N (wt. %)	0.8
S (wt. %)	2.5
O (wt. %)*	4.7
Proximate analysis	
Volatile (wt. %)	65.1
Fixed carbon (wt. %)	29.9
Ash (wt. %)	4.9
Chemical composition (wt. %)	49.6
Natural rubber (NR)	32.1
Synthetic Butadiene Rubber (SBR) Additives	18.3
Moisture (wt. %)	0.6
HHV (MJ/kg)	38.2

a. * by difference

b. ** on dry basis

c. *** on a carbon black and free of ash basis

In pyrolysis, the heat decomposition of biomass involves a complex interaction between the heat balance and the mass balance, which are several chemical reactions that produce condensable vapor (bio-oil), product gas (pyro-gas), and solid charcoal (bio-char) [20]. In this study, the pyrolysis process used temperatures of 400-600 °C and was carried out in a batch reactor for 1 hour. The process occurs in atmospheric conditions by utilizing variations in the temperature. The reactor used is shown in Fig. 3. The batch reactor is made of stainless steel and uses a hot stove as the heater needed to produce the liquid product from the process. The pyrolysis process took place at temperatures of 400, 450, 500, 550, and 600 °C without using a catalyst.

3.2. Effect of Temperature on Product Yield

In the pyrolysis process, the resulting product's most affected is the operating temperature used. The higher the temperature used in the pyrolysis process, the higher the gas product produced, and the lower the operating temperature used, the dominant products produced are solids and liquids. The effect of temperature on the product yield of the pyrolysis reaction is quite significant. The increase in operating temperature causes a decrease in the yield of carbon black. In contrast, the yield of gaseous products always increases with increasing operating temperature, and the yield of liquid products initially increases with increasing operating temperature and then decreases with a further increase in temperature [21]. The steam produced by the reactor is then liquefied with the help of a condenser. During the pyrolysis process, large molecules of liquid tires are converted into steam and exit the reactor. The steam produced by the reactor is then liquefied with the help of a condenser. The pyrolysis reaction produces the first liquid at an operating temperature of 400 °C [22]. The most important parameter affecting the pyrolysis reaction process is the temperature. Maximum liquid product yield is reached at 500 °C [23]. The yield of the resulting liquid product increases with increasing operating temperature in the pyrolysis process as presented in Table 2.

Table 2. Yield (% wt.) relationship to temperature (°C).

Materials	Temperatures (°C)				
	400	450	500	550	600
Raw Materials (g)	2000	2000	2000	2000	2000
Liquid Yields (% wt.)	39.32	39.02	42.58	42.11	41.84
Solid Yields (% wt.)	40.73	41.54	38.82	37.68	35.91

From Table 2. It can be seen that the liquid product increased with the increasing temperature used in the pyrolysis process. The highest yield of liquid products obtained at a temperature of 500

°C is 42.58 %wt, and the lowest is at a temperature of 450 °C, which is 39.02 %wt. The highest yield of solid products obtained at a temperature of 450 is 41.54 %wt, and the lowest is obtained at a temperature of 600 °C, namely 35.91 %wt. In the pyrolysis process of waste tires without using a catalyst, the liquid yield at temperatures of 400, 450, 500, 550, and 600 °C, respectively; 39.02; 42.58; 42.11; and 41.84 %wt. The yield of solids produced at temperatures 400, 450, 500, 550, and 600 °C, respectively, was 40.73; 41.54; 38.82; 37.68; dan 35.91 and 35.91 % wt as shown in Fig. 5.

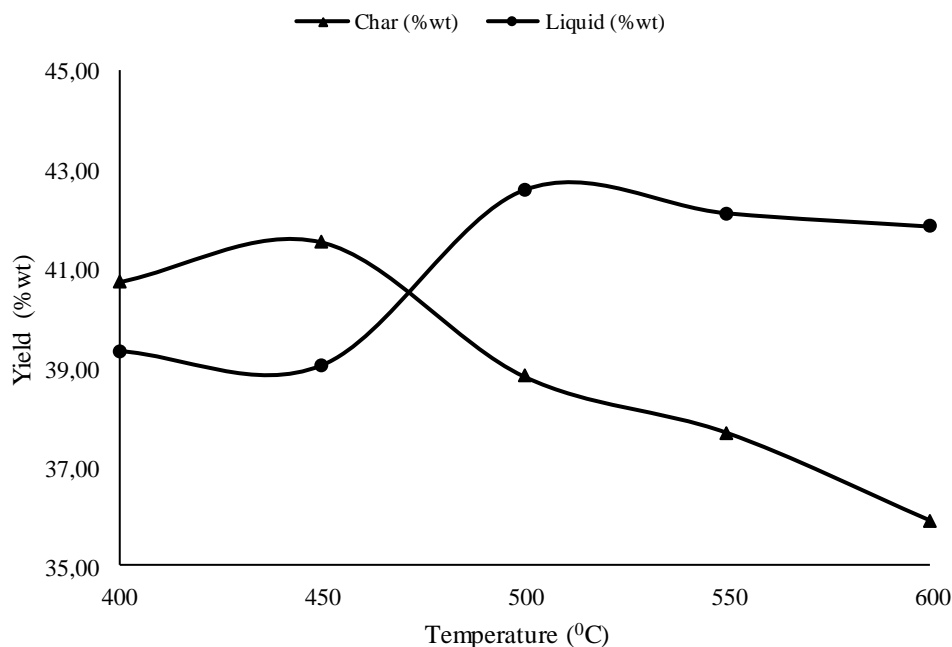


Fig. 5. Product yield (%wt) at Various Temperatures.

Fig. 5 shows the product yield from the pyrolysis process of used tire waste. The liquid yield obtained at a temperature of 400 °C is 39.2 % wt, then at a temperature of 450 °C a slight decrease is obtained; the yield is 39.02 %wt. The decrease that occurs is insignificant, so this does not affect the results obtained at increasing temperatures. Then the yield increased at a temperature of 500 °C, which is 42.58 %wt. Then at a temperature of 550 and 600 °C, the yield obtained decreased. This happens because the most optimal yield is obtained at a temperature of 500 °C, at a temperature of 550 and 600 °C, there is no increase. The yield of solids and liquids at temperatures of 550 and 600 °C decreased because, at these temperatures, the products that increased were gas products. The maximum yield of liquid is obtained at the reaction temperature of 500 °C. Operating temperatures below 500 °C will produce more solids, and above 500 °C will produce more gases [24]. It can be seen that at temperatures of 550 and 600 °C the yield of solids and liquids decreased, so it can be identified that the gaseous products increased at these temperatures. These data obtained the most optimal liquid at a temperature of 500 °C.

In this study, the product obtained is liquid in the form of concentrated black oil, almost similar to petroleum. To take advantage of these liquid products, further processing is needed to obtain the desired diesel oil. The advanced process can use the distillation process to separate the impurities in the product so that the desired results can be obtained. The results of this research need to be analyzed further to obtain the characterization of the liquid product obtained. Optimization of the reactor and adding tools are required for further research, so that the resulting product characterization can be obtained.

4. Conclusion

In the research that has been done, two types of products can be identified directly, namely liquid products and solid products. The liquid product increases as the operating temperature increases in the pyrolysis process. The highest liquid product yield was obtained at a temperature of 500 °C, 42.58 % wt, and the lowest liquid product yield was obtained at a temperature of 450, 39.02 % wt. The overall liquid product obtained at temperatures 400, 450, 500, 550, and 600 °C is 39.2, 39.02, 42.58, 42.11, and 41.8 % wt, so the most optimal temperature for producing liquid product is 500 °C. The solid product decreased with increasing operating temperature. The solid products obtained at temperatures 400, 450, 500, 550, and 600 °C are 40.73, 41.54, 38.82, 37.68, and 35.91 wt%, respectively. Solid products are also very influential on the operating temperature used.

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