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Production and Characterization of Liquid Smoke from Coconut Shell Waste as an Effort to Reduce the Impact on Environmental Pollution

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ABSTRACT

This research examined the influence of pyrolysis temperature on a large-size feedstock of coconut shell waste for producing biochar and liquid smoke using slow pyrolysis. The temperature used was varied between 250 °C to 450 °C at a constant heating rate of 10 °C/min and at a holding time of about 120 minutes. Gravimetry, spectro, and high-performance liquid chromatography (HPLC) methods are used to identify the liquid smoke for density, phenol, and acetic acid content respectively. The results indicated that increased pyrolysis temperatures caused a reduced biochar yield. However, the liquid smoke yield was found to increase with the temperature. The obtained liquid smoke has a density of 1.054 g/mL and has a content of phenol of about 4717.91 mg GAE/100 mg, and acetic acid of 11.36%. Results inferred that the liquid smoke can be generated from a large size of coconut shell through pyrolysis at medium temperature.

Keywords: coconut shell, pyrolysis, liquid smoke, biochar.

INTRODUCTION

Biomass waste, such as agricultural, food, or wood, can have a significant environmental impact if not managed properly. According to Roy et al. [2014], when biomass waste decomposes naturally or is burned incompletely, such as in open waste burning or biomass burning for energy, it can produce greenhouse gas emissions such as carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). Burning biomass waste under uncontrolled or inefficient conditions can produce emissions of particulates, air pollutants such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), and volatile organic compounds (VOCs) also a serious threat to the environment as revealed by Demirbas [2007]. Soil and water also experienced degradation [Bhattacharyya et al., 2015]. Another threat is the deterioration of water quality [Akhtar et al., 2021], and biodiversity reduction [Dauber et al., 2010]. Iakovou [2010] states that inefficiently disposed biomass waste represents a potential loss of valuable resources.

To address the environmental issues associated with biomass waste, it is essential to implement sustainable and environmentally friendly waste management practices, such as recycling, composting, or the use of advanced waste treatment technologies as well as waste-to-energy technology. However, combustion-based biomass waste-to-energy technology is still developing. The study and development of incinerators to produce thermal energy and low emissions continue [Baharuddin et al., 2022; Simanjuntak, et al., 2022]. In addition, it is also necessary to encourage the adoption of technologies and policies that support the use of biomass waste as well as coconut shells as a valuable source of phytochemicals, which can be converted into value-added products. Coconut shells are an abundant agricultural waste due to the large amount of coconut production in various tropical regions including Indonesia [Alouw and Wulandari, 2020]. This waste is often not fully utilized and is often disposed of or incinerated, causing environmental problems. Coconut shells have great potential to produce raw material as liquid smoke production [Singh et al., 2024]. Using coconut shells as raw material as liquid smoke can turn waste into a valuable resource. The coconut shell contains complex organic compounds that can be converted into liquid smoke. These compounds include carbohydrates, lignin, and cellulose that can decompose into products such as phenols, aromatic hydrocarbons, and other oxygen compounds that contribute to the composition of liquid smoke [Rizal et al., 2020]. In addition, the coconut shell has a large enough surface area, allowing a higher yield of liquid smoke.

The liquid smoke obtained from coconut shells has various uses in various fields. It can be an alternative fuel to replace conventional fossil fuels such as gasoline or diesel to reduce dependence on fossil energy sources and greenhouse gas emissions [Hossain and Davies, 2013]. Coconut shell liquid smoke can also be used as an additive to improve the quality of conventional fuels [Gunasekar et al., 2021]. Adding liquid smoke to fuels can improve calorific value, stability, and combustion performance and reduce emissions of pollutants such as nitrogen oxides (NO_x) and particulates. Coconut shell liquid smoke can be used as a raw material in the chemical industry to synthesize various chemical products such as phenol, acetic acid, and aromatic hydrocarbons [Mulyawanti et al., 2019].

Coconut shell liquid smoke can be used in the food and beverage industry as an additive to impart a smoked taste and aroma to products such as meat, fish, cheese, sauces, and beverages. It provides a natural alternative to provide a smoky flavor without the need to use traditional smoking processes [Sari et al., 2019]. Some compounds in coconut shell liquid smoke can also be used in the cosmetic industry and personal care products as additives to provide aroma and enhance the quality of products such as soaps, creams, or shampoos [Nurfadhila and Hambali, 2022]. Coconut shell liquid smoke can be used as a natural wood preservative which can help protect it from fungal attacks, pests, and degradation due to environmental factors [Kailaku et al., 2017]. Coconut shell liquid smoke can also be used as a liquid fertilizer or additive to improve soil fertility and agricultural productivity [Diptaningsari et al., 2022].

Liquid smoke is generally obtained through pyrolysis, a thermal process in which coconut shell is heated in a low-oxygen environment. Until now, this technology continues to be developed to explore the added value of coconut shell waste [Chan et al., 2022]. Depending on the intended use, pyrolysis can be carried out on a laboratory, pilot, or industrial scale. Pyrolysis temperature can vary depending on the type of raw material and desired process conditions but generally ranges from 300 °C to 600 °C [Liu et al., 2015]. The raw material undergoes thermal decomposition during heating, producing various thermal products, including smoke, gas, and biochar. The resulting smoke will condense into liquid smoke during cooling. The smoke produced from the pyrolysis process is then condensed to form liquid smoke. This is usually done by cooling the pyrolysis vapor so that the smoke condenses and condenses into a liquid form.

It is important to note that the coconut shell pyrolysis process must be carried out carefully to ensure the sustainability and efficiency of the process and take into account the environmental impact of the production and use of liquid smoke. The coconut shell's size significantly influences the production of liquid smoke because it dramatically affects the heat conductivity in the material. The smaller the size of the coconut shell, the greater its surface area is exposed to heat, allowing heat dispersal throughout the particles to occur more efficiently. The smaller coconut shell tends to have higher heat conductivity, so it can produce higher and more even temperature during the pyrolysis process, improving the conversion efficiency of the material into liquid smoke.

Many researchers have conducted pyrolysis of coconut shells on small particle sizes from millimeters (mm) to micrometers (µm) [Septien et al., 2012; Luo et al., 2010; Suriapparao and Vinu, 2018; Zhang et al., 2017a]. All of them conclude that particle size greatly affects liquid smoke products. Selecting the right coconut shell size is very important to maximize the production of liquid smoke during the pyrolysis process. Although the small size of the coconut shell can provide some advantages in the pyrolysis process, some difficulties can arise related to using small coconut shells. Crushing coconut shells into smaller sizes requires additional crushing or grinding processes. Too small size can also lead to significant energy losses due to the processing process and manipulation of raw materials. This process requires additional time, labor, and cost to prepare the raw materials before the pyrolysis process can begin. Processing small coconut shells can produce dust and particulates, which can be a hygiene and health problem for workers. This dust can irritate the respiratory tract; additional measures are required for dust control and waste management. The small size of the coconut shell can also increase the risk of environmental pollution, especially if there are no proper pollution control measures in place. Dust and particulates produced during treatment can be dispersed into the air, soil, and water, negatively impacting the surrounding environment.

However, the large size of the coconut shell can also provide some advantages in producing liquid smoke through pyrolysis. The large coconut shell tends to have better temperature stability during pyrolysis. This is because the larger mass can store heat more efficiently, so the temperature inside the reactor is more consistent and accessible to regulate. The large size of the coconut shell allows easier control of the pyrolysis process. This provides greater flexibility in regulating temperature, time, and other parameters to produce liquid smoke of the desired quality. Large coconut shells tend to produce less dust and particulates during processing. This can reduce the risk of environmental pollution and worker health problems associated with dust and particulates.

Using large coconut shells can improve processing efficiency by minimizing the time and labor required for crushing and processing raw materials. This can reduce operational costs and increase overall process productivity. Although a coconut shell's large surface area may be lower than a coconut shell crushed into a small size, the pyrolysis process can proceed more efficiently and produce a higher liquid smoke yield due to better temperature stability. However, it should be noted that using large coconut shells also has some disadvantages, such as longer processing times and difficulties in achieving efficient separation and condensation. Therefore, selecting the optimal coconut shell size must consider various factors, including process efficiency, product quality, and production costs. Therefore, large-scale liquid smoke production could be more efficient using small particle sizes. Pyrolysis studies on coconut shells of large or pebble sizes are necessary. So far, there has been no study of the pyrolysis of large coconut shells above several centimeters (cm). This research examines coconut shell pyrolysis with a large size. Pyrolysis is carried out at several operational temperatures to obtain an optimal liquid smoke product.

MATERIALS AND METHOD

Material

The coconut shell was used as the feedstock in this study. It was obtained from a traditional market around the research location. The feedstock form and appearance as depicted in Figure 1. After chopping to an average of 5 × 5 cm, the pre-drying treatment was conducted using sunlight for several days. This step was important to reduce the moisture content for more efficiency during the heating process.

Previous studies have indicated the proximate and ultimate characteristics of coconut shells as shown in Table 1. As coconut shell has high volatile, low ash, and fixed carbon, there is a higher possibility for coconut shells to be converted into biochar and liquid smoke.

Procedure

The schematic diagram of the experiment is shown in Figure 2. The feedstock was manually filled into the stainless steel pyrolyzer. About 3 kg of feedstock was used for each pyrolysis run. The selected operating parameters in this study include a heating rate of 10 °C/min and a holding time of 120 minutes. This holding time is

Figure 1. Pebble size feedstock sample used

Table 1. Ultimate and proximate average value of coconut shell [Ganapathy Sundaram and Natarajan, 2009]

Proximate and ultimate analysis of coconut shell								
Elemental analysis (wt.%)					Proximate analysis (wt.%)			
C	H	N	O	S	Moisture	Volatile	Fixed carbon	Ash
58.73	6.15	0.54	38.45	0.02	6.98	72.93	19.48	0.61

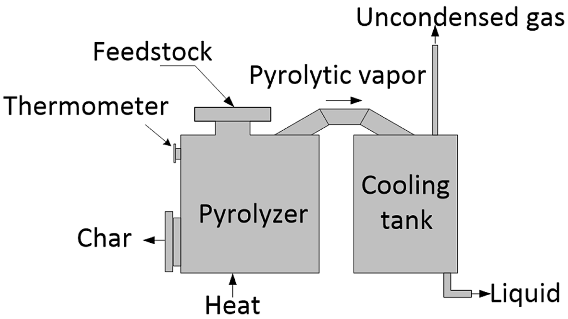

Figure 2. Schematic of the pyrolysis system in this study

sufficient because the main purpose is to produce liquid. According to Saker [2018], the effective holding time to produce liquid from coconut shell pyrolysis is the first 20 minutes at its optimum temperature. Thus, the only parameter studied is temperature as the most important parameter in the pyrolysis process [Zhang et al., 2017b]. Temperature ranges from 250 °C to 450 °C were applied to identify the optimal operating temperature for achieving maximum yields. The initial temperature of 250 °C was chosen because, at this point, the carbonization process initiates [Ahmad et al., 2020]. The weight of the product was determined by weighing it after each pyrolysis run. For each pyrolysis temperature, the average product yields from five pyrolysis runs were presented. The yields and heating values were calculated using Eqs. 1 and 2 and expressed in weight percent (wt.%).

$$\text{Liquid yeild } \% = \frac{\text{total liquid product weight}}{\text{feed weight}} \times 100 \quad (1)$$

$$\text{Char yeild } \% = \frac{\text{char weight}}{\text{feed weight}} \times 100 \quad (2)$$

RESULTS AND DISCUSSION

Pyrolysis production

Figure 3 depicts the physical characteristics of pyrolysis products obtained from coconut shells. In its liquid and charcoal (in solid form) exhibit a distinct dark color (Kan et al., 2016a). Liquids derived from coconut shells typically display specific features, including a high oxygen content, elevated water content, lower pH, and a low heating value. Notably, the water content

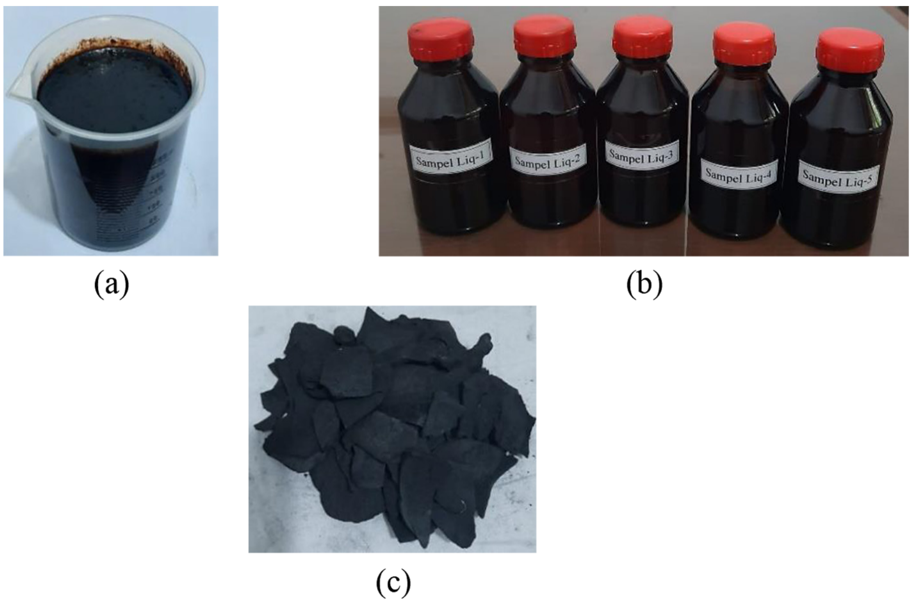

Figure 3. Physical product sample: (a) liquid, (b) liquid sample for the characteristic test, (c) biochar

becomes a critical concern in the bio-oil production process, posing a significant challenge to its efficient use as a fuel. The substantial presence of oxygen in bio-oil is primarily attributed to the existence of oxygenated compounds. As cellulose and hemicellulose are the main constituents of coconut shells, the resulting bio-oil is predominantly composed of acids and ketones. Addressing these specific attributes is crucial for researchers and engineers seeking to optimize bio-oil production processes and enhance their applicability for various purposes. Both products depend on the temperature of the process carried out. Just like the studies that have been done char products decrease with temperature increase, while liquid increases with temperature to a certain optimum temperature [Ahmad et al., 2020].

Figure 4 illustrates the pyrolysis yields at various temperatures. It is observed that the biochar decreases with increasing temperature, while the liquid smoke shows an increase with rising temperature. As the pyrolysis temperature ranges from 250 °C to 450 °C, the liquid yield initially rises, reaching its peak at a temperature of 400 °C and decreases when the temperature is increased. According to Gao [2016], at higher temperatures, carbonization and the production of non-condensable gases are thought to primarily occur, resulting in a decrease in condensable products. The breakdown of coconut oil into small-molecular-weight non-condensable gas hydrocarbons or hydrogen reduces the yield of coconut oil.

The study determined that the optimal liquid was obtained at a temperature of approximately 400 °C, aligning with the findings of previous research by Demirbas [2004]; [Rout et al., 2016]; [Sarkar and Wang, 2020]. This could be attributed to nearing the completion of the carbonization stage at that temperature. The decision to use a feedstock size of 5 cm² was informed by the study of Hasan [2022] where this size resulted in biochar with a fully black color compared to a size of 7 cm². Similar results were obtained by another researcher using feedstock sizes of 2 cm [Kan et al., 2016b].

Liquid smoke characteristic

Coconut shells contain a wide variety of organic components, including cellulose, hemicellulose, lignin, and various other compounds. During the pyrolysis process, the high temperature applied causes thermal decomposition of these components. In this work, the main components of liquid smoke are phenolic and acetic acid. Gravimetry, spectro, and HPLC methods are used to measure the characteristics of liquid smoke for density, phenol content, and acetic acid content, respectively. Fig. 5 shows the effect of temperature on the density, phenol production, and acid content of coconut shell pyrolysis. Density is not too influenced by temperature, while the acid content is found to be optimal at 300–350 °C, followed by the phenol content obtained optimally at 350 °C.

The liquid smoke produced has products similar to those obtained with the same pyrolysis temperature parameters by Dewi [2022], which are dominated by acetic acid and phenol, which are very important in agriculture as antioxidants, antiseptics, and antibacterial agents. Another study using large feedstock sizes also managed to get liquid smoke from corncobs. They concluded that there would be a decrease in liquid smoke production if the size was increased. There is a 6.5% decrease in liquid smoke production if the particle

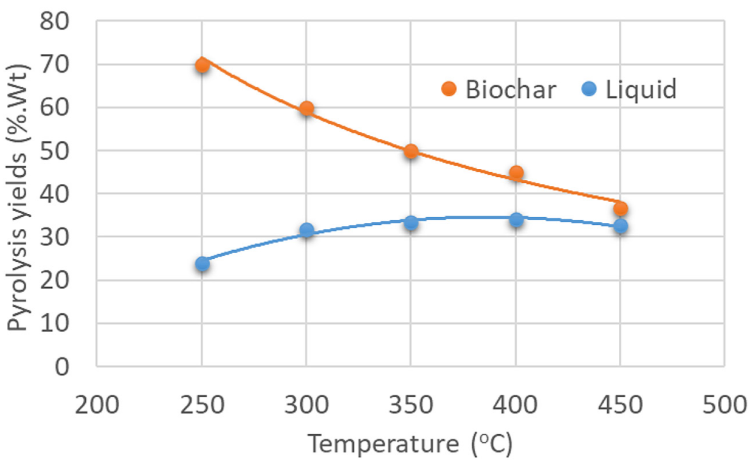

Figure 4. Product yield at different temperatures

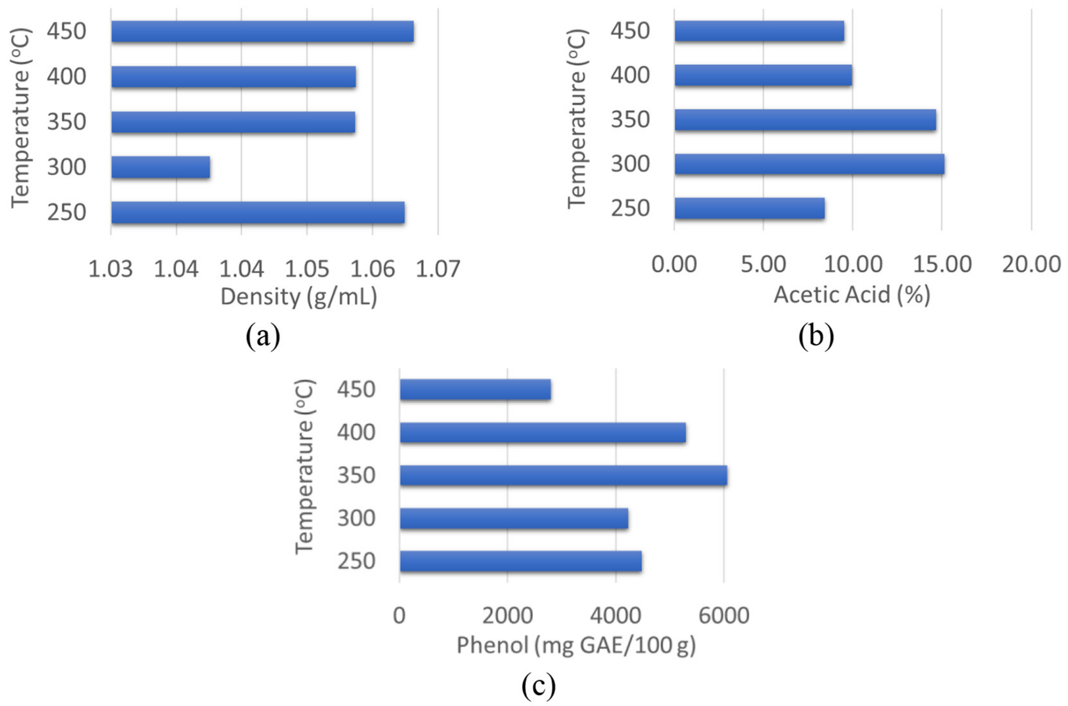

Figure 5. Effect of temperature on pyrolytic compound

size is increased from 0.375 cm to 3.75 cm [Aladin et al., 2018]. Compared to the percentage increase in particle size (90%), the decrease in liquid smoke production is considered slight. In addition, indeed, making particle size requires high costs.

Figure 5 describes that acetic acid and phenol were identified as the largest compounds contained in liquid smoke. This finding agrees with research conducted by Adinda [2023], where they obtained liquid smoke from coconut shells with dominant acetic acid and phenol content. Acetic acid is obtained from the hemicellulose degradation process, while phenol is obtained from the lignin degradation process. Phenol and acetic acid are important components of liquid smoke due to their useful applications. Phenols have antioxidant, antiseptic, and antibacterial properties. Meanwhile, acetic acid can exhibit antimicrobial activity and can also determine taste and flavor. Figures 5b and 5c show that the content of acetic acid and phenol decreases with temperature. This decrease was possibly due to the fact that the liquid smoke was further decomposed into other compounds beyond 350 °C. This finding is in accordance with the results found by Faisal [2018].

Related to the size of the feedstock, which is crucial for liquid smoke production, this research is also supported by data and the fact that a large feedstock does not greatly affect pyrolysis results. Bamboo material with a size of 5 cm can produce liquid smoke in competition with other biomass with small particle sizes [Swandewi et al., 2019]. Other researchers were also able to obtain liquid smoke from coconut shells with a particle size of 2–3 cm [Jahiding et al., 2015] with temperature parameters similar to those in this study. They found that acetic acid and phenol content were the most dominant ingredients in liquid smoke. The same is done by Gao et al. [2016]. By testing the particle size of feedstock, they proved that the smallest particle size is not the main parameter in producing liquid smoke. However, because it has a higher amount of phenolic compounds and a smaller proportion of pentane, aldehyde, and furans, as well as a higher proportion of acetic acid and nitrogen-containing chemicals, coconut shells are not a viable source of liquid fuel. Rather, it can serve as a valuable supply of gaseous fuel like syngas.

CONCLUSIONS

In this work, the thermal decomposition of coconut shells to produce liquid smoke and biochar via the pyrolysis process was studied. Pebble sizes of feedstock are hoped to be the exact choice for the coconut shell pyrolysis. Coconut shells feedstock at an average size of 5 cm^2 was used and the temperature ranges of 250 °C to 450 °C were performed to get the optimum yield of biochar and liquid smoke. From the experimental evaluation, a temperature of 400 °C and a residence time of 120 minutes stand to be recommended. More studies on the characteristics of charcoal are required to recommend the best production process operating conditions with significant improvement of biochar. The findings of the present work indicate that the quality properties of liquid smoke might be obtained at large sizes of feedstock. Another work could be performed to study pyrolysis on a bigger size than this work.

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