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Hybrid Biopellets Characterization of Gamal Wood (*Gliricidia sepium***) and Robusta Coffee Husk at Various Compositions**

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ABSTRACT

Biopellets from gamal wood (*Gliricidia sepium*) as a biomass energy resource could be an alternative to replace fossil fuels due to having met standards based on moisture content, ash content, fixed carbon, calorific value, and density. Unfortunately, they still had high levels of volatile matter. Robusta coffee husk was a material with high nitrogen content, which is suspected of being able to bind aromatic substances in volatile organic compounds. This study aims to evaluate the quality of biopellets and determine the optimum composition of the biopellets from gamal wood and coffee husk. The blended composition of gamal wood and coffee husk biopellets studied were 100:0, 75:25, 50:50, 25:75, and 0:100. The biopellets were manufactured using the material size of 40-60 mesh with a pressure of 173.51 MPa. The best biopellet was produced in the composition of 75% gamal wood and 25% coffee husk, with a density of 0.85 g/cm³, moisture content of 8.03%, ash content of 3.92%, volatile substances of 78.01%, fixed carbon of 18.07%, and calorific value of 4,176 cal/g. The biopellet quality met the standards of SNI 8021:2014 and EN 14061-2, except for ash content. Adding coffee husk reduced gamal wood biopellet's volatile matter, increasing the fixed carbon and density of gamal wood biopellets.

1. Introduction

In these days, renewable energy sources such as biomass are widely developed. Biomass can be obtained from plants and animals. BPS (2023) reported that biomass had a large energy input source in Indonesia from 2017 to 2021. Biomass contributed 526 petajoules (PJ) of energy input in 2017, which increased in 2018 and 2019 to 640 PJ and 802 PJ, respectively. The increase in energy input also occurred from 2020 at 774 PJ to 799 PJ in 2021. The significant energy input potential makes biomass an energy source that can be developed as an alternative to fossil fuels. In addition to the enormous energy input potential, biomass energy was also more environmentally friendly regarding emissions. Biomass as an energy source in the gasification process could reduce carbon dioxide emissions by 3–11% (Pramudiyanto and Suedy 2020). On the other hand, coal produces 94% higher greenhouse gas emissions compared to wood-based biopellets (Wang et al.

2017). However, biomass had unfavorable disadvantages compared to coal, such as large volume, low density, low energy value, hygroscopicity, high moisture content, and high variability when used as an alternative energy source (Chen et al. 2021; Yang et al. 2019). One way to overcome the biomass's variability in form and size was by converting biomass into biopellets.

Biopellets were one of the biomass energy sources that had the potential to be developed. Biopellets were produced by densifying biomass to improve their physical, mechanical, and calorific properties (Wahyono et al. 2021). Biopellets could be made from wood, agricultural waste, and other lignocellulosic materials. Wood-based biopellets had massive potential criteria to become an alternative to fossil fuels, such as economic value, enormous raw materials, import competition, technology, the environment, community, and policy (Rimantho et al. 2023). Woodbased biopellets do not require high-quality wood, so the wood usually used is low-value-added (Goetzl 2015). Fast-growing wood is a potential source of biopellet feedstock that can be developed. One of the fast-growing woods in Indonesia with superior potential is gamal wood (*Gliricidia sepium*).

Gamal in the form of firewood is estimated to supply the energy needs of a power plant of 18.90 megawatts (Hudaedi et al. 2018). Gamal is a fast-growing species likely to grow naturally without complex maintenance (Atapattu et al. 2017). Maulana et al. (2021) reported that gamal was one of the potential biomass energy resources cultivated by the local community and usually used to be barn feedstock, nectar resource, and soil conservation. Biopellets from gamal wood have high levels of volatile matter (Rahayu et al. 2022). Therefore, it is necessary to improve the quality of gamal wood pellets by reducing the high levels of volatile matter. The composition of the raw materials for making biopellets determines the levels of these volatile substances (da Silva et al. 2020). One of the potential ingredients to be mixed in gamal wood biopellets is coffee husks.

Coffee husks were the leading waste from coffee processing, representing 12 wt % of coffee grains (Manrique et al. 2019), especially for robusta coffee husks in Lampung Province. Robusta coffee production in Lampung Province reached 131,501 tons in 2014 (BPS 2022). Based on its abundant availability, the coffee husk can be made into more valuable products, including biopellets. Coffee has a high nitrogen content of 2.6% (Cruz et al. 2012), compared to pine wood, which only has a nitrogen content of 0.117–0.547% (Köster et al. 2015). Nitrogen can bind aromatic compounds (Rinawati et al. 2019), which are part of the volatile organic compounds in wood (Fang et al. 2014). Nugraha et al. (2020) reported that robusta coffee contained nitrogen compounds that could induce carbon to absorb odor or aromatic volatile matter.

Biopellets from gamal wood had a moisture content of 3.00–5.62%, density of 1.25–1.30 g/cm³, ash content of 1.11–1.20%, volatile matter of 80.30–82.96%, fixed carbon of 10.31– 18.40%, and calorific value of 4,351–4,494 Kcal/kg (Rahayu et al. 2022; Gunawan et al. 2023). The proximate values met the SNI 8021:2014 except for the volatile matter, which had to be less than 80% (BSN 2014). Therefore, mixing coffee husks needs to be done because it was thought to reduce the volatile matter value of gamal wood biopellets due to the nitrogen compounds in coffee husks that could bind the aromatic compounds.

Based on the previous explanations, a study was conducted on the manufacture and quality testing of biopellets mixed with gamal wood and coffee husk based on SNI 8021:2014 (BSN 2014) and EN 14961-2 (European Committee for Standardization 2011). In addition, this study aimed to determine the best material composition in biopellets mixed with gamal wood and coffee husk. The composition of hybrid biopellets used in this study was with the ratio of gamal wood and coffee husk of 100:0, 75:25, 50:50, 25:75, and 0:100.

2. Materials and Methods

2.1. Materials

The materials used in this research were gamal wood (*Gliricidia sepium*) and robusta coffee husk. Gamal wood came from the "Taman Hutan Raya (Tahura) Wan Abdul Rachman", H5JP+425, Pesawaran Regency, Lampung 35156, in the utilization block. In conservation areas, the local community could use the utilization block to support their economic welfare (Erwin et al. 2017). Robusta coffee husks were obtained from the coffee milling factory, JVWH+33X, Lugusari, Pagelaran, Pringsewu Regency, Lampung 35376.

2.2. Material Preparation

Gamal wood, still in log form, was chopped using a machete to produce wood chips. Gamal wood chips and coffee husks were sun-dried until less than 12% moisture content was reached. The woods were ground using a hammer diesel engine, and the coffee husks were ground using a hammer herb grinder. The resulting gamal wood powder and coffee husks were then filtered with a sieve shaker stuck on a 40–60 mesh sieve.

2.3. Biopellet Manufacturing

Gamal wood powder and coffee husk powder that had been obtained were then weighed and mixed as much as 3 g with the ratio of gamal wood powders (G) and coffee husk powders (C) as much as 100:0 (G100C0), 75:25 (G75C25), 50:50 (G50C50), 25:75 (G25C75), and 0:100 (G0C100). Then, the powders were put into a pellet mold with a diameter of 1.2 cm and pressed using a hydraulic press with a pressure of 173.51 megapascals (MPa). An illustration of the biopellet manufacturing process is presented in **Fig. 1**.

Fig. 1. Illustration of the biopellet manufacturing process.

2.4. Biopellet Characterization

Characterization of biopellets referring to SNI 8021: 2014 and EN 14961-2 concerning Quality Requirements for Pellets is shown in **Table 1**.

2.4.1. Density determination

The weight of the sample was measured, and the diameter and the longest part of the pellets in the center were measured using an analytical caliper. The density of each test sample was calculated using Equation 1.

$$
\rho\left(g/cm^3\right) = \frac{m}{V} \tag{1}
$$

where ρ is the density of the biopellet (g/cm³), *m* is the sample weight (g), and *V* is the sample volume $\text{(cm}^3)$.

Parameters	Unit	SNI 8021:2014	EN 14961-2		
			ENplus-A1	ENplus-A2	EN-B
Density	g/cm^3	≥ 0.8		≥ 0.6	
Moisture content	$\frac{0}{0}$	≤ 12		≤ 10	
Ash content	$\frac{0}{0}$	≤ 1.5	≤ 0.7	≤ 1.5	\leq 3
Volatile matter	$\frac{0}{0}$	≤ 80		$\overline{}$	
Fixed carbon	$\frac{0}{0}$	≥ 14		۰	
Calorific value	cal/g	$\geq 4,000$	$\geq 3,940$	$\geq 3,890$	$\geq 3,820$

Table 1. Biopellet quality requirements (BSN 2014; European Committee for Standardization 2011)

2.4.2. Moisture content determination

The biopellets were placed in a porcelain cup whose weight was already known, and then the sample was placed in an oven at 103±5°C for 3 hours until the mass was constant. The biopellets were conditioned in a desiccator until the condition was balanced. The moisture content was calculated using Equation 2.

$$
Moisture content (%) = \frac{W_0 - W_1}{W_1} \times 100
$$
\n(2)

where W_l is the weight of the sample before drying in the oven (g), and W_0 is the oven-dried weight of the sample (g).

2.4.3. Ash content determination

Samples of 1*–*2 g were placed on a porcelain cup of known weight, and then the samples were placed in a furnace at 650°C for 5 hours. The samples were then cooled in a desiccator until stable and balanced. The calculation of ash content was done using Equation 3.

$$
Ash content (%) = \frac{W_a}{W_0} \times 100 \tag{3}
$$

where W_a is the ash mass (g), and W_0 is the mass of the sample before being placed in a furnace (g).

2.4.4. Volatile matter determination

2 g of oven-dried samples were placed on a porcelain cup whose known weight. Then, the sample was put into the furnace at 950°C for 7 minutes. Then, the sample was put into a desiccator until balanced conditions. The volatile matter was calculated using Equation 4.

$$
Vola tile matter (%) = \frac{W_1 - W_2}{W_1} \times 100 \tag{4}
$$

where $W₁$ is the oven-dried sample (g), and $W₂$ is the mass of the sample after heating in the furnace (g).

2.4.5. Fixed carbon determination

Fixed carbon was calculated using Equation 5, following the SNI 8021 (BSN 2014).

$$
Fixed carbon (%) = 100 - (A+B)
$$
\n
$$
(5)
$$

where *A* is the ash content $(\%)$, and *B* is the volatile matter $(\%)$.

2.4.6. Calorific value determination

1 g of the sample was crushed, placed in a silica cup, and tied with a 10 cm nickel wire. After that, the test sample was put into a tube, closed tightly, and supplied with 25 atm of oxygen. The tube is inserted into an oxygen bomb calorimeter (plain jacket calorimeter Cat. 1341CLEE made by Parr Instrument Company, USA) filled with water. The temperature rise is observed every one minute until it reaches the optimum temperature. The calorific value was calculated using the Equation 6.

Calorific value (cal/g) =
$$
\frac{(T.V - e_1 - e_2 - e_3)}{m}
$$

\n(6)

where *T* is the temperature rise on the thermometer $({}^{\circ}C)$, *V* is the calorimeter energy equivalent determined based on standardization (2,426 calories/°C), *e1* is ml of sodium carbonate used for titration (23.9 calories), e_2 is $13.7 \times 1.02 \times$ sample weight, e_3 is $2.3 \times$ length of burnt nickel wire, and *m* is the mass of biopellet (g).

2.5. Fourier Transform Infrared Spectroscopy (FTIR) Analysis

5 mg of biopellet powders (40*–*60 mesh) were analyzed using an FTIR spectrometer (Bruker-Tensor II, Germany) over the range 4000–500 cm-1 by the Attenuated Total Reflectance (ATR) method. Samples were scanned 45 times with a resolution of 2 cm.

2.6. Data Analysis

This study's experimental design was a simple, completely randomized design (CRD) with a single factor, such as heat treatment. The data obtained in this research were analyzed statistically using analysis of variance (ANOVA) followed by Duncan's Multiple Range Test (DMRT) to determine a significant difference within the factor's level. The statistical analysis was determined using International Business Machines (IBM) SPSS Statistics Software 22.

3. Results and Discussion

3.1. Density

The density of biopellets of all variations of material composition is shown in **Fig. 2**. The result shows that the biopellets density ranged from $0.838-0.929$ g/cm³, with the highest density found in the G0C100 composition and the lowest density found in the G100C0 composition. Based on the results of the analysis of variance ($\alpha = 0.05$), it shows that the addition of robusta coffee husk into gamal wood biopellets has a very significant effect ($p \le 0.01$) on the value of biopellets density.

Fig. 2. The density of biopellets with various composition variations; different letters indicate significantly different results.

The density of biopellets of all variations of material composition has reached the standards of SNI 8021: 2014 and EN 14961-2. The coffee husk powders and gamal wood powders examined had a particle size of 40–60 mesh. With the same particle size, it is suspected that coffee husk powder has a higher percentage of smaller particle sizes than gamal wood powder. This result causes the coffee husk powders to be finer than the gamal wood powder. Rebolledo-Hernández et al. (2023) reported that the particle size of coffee husk was $\leq 250 \,\mu$ m, causing strong inter-particle forces so that coffee powder was more cohesive. This phenomenon caused the biopellets made from gamal wood and coffee husk to have a higher density value than biopellets from gamal wood alone. Likewise, Budiawan et al. (2014) also reported that coffee husk particles tend to be finer than wood powder particles, thus minimizing the space between the particles. Biopellet density had to be high due to the combustion reactivity and quality. Wang et al. (2023) reported that characters resulting from the pyrolysis of pellets of higher density exhibit a higher oxygen content in conjunction with a reduced concentration of crystallite clusters, thereby rendering them more reactive in combustion tests.

3.2. Moisture Content

The analysis result showed that the moisture content of the biopellets studied ranged from 7.871%–8.487%, with the highest moisture content found in the G0C100 composition and the lowest value found in the G100C0 composition. The moisture content of biopellets of all material composition variations is presented in **Fig. 3**.

The variance results showed that adding coffee husk into gamal wood biopellets had a very significant effect ($p \le 0.01$) on the moisture content of biopellets. Overall, the moisture content of biopellets has reached the target of SNI 8021:2014 and EN 14961-2. The moisture content was an important property in biopellets due to its impact on the calorific value. The moisture content of biopellets had to be low. Miranda et al. (2015) reported that high moisture content in biopellets indicated that combustion heat was employed in the partial water evaporation from the fuel. In contrast, for dry biopellets, the entire heat was utilized for the intended purpose. Lower moisture contents facilitate elevated flame temperatures, which in turn result in enhanced temperature gradients and heat transfer, thus enabling the completion of combustion. The addition of coffee

husk can increase the moisture content of biopellets. In another study, coffee husk powder was reported to have a moisture content of 8.97% (Rebolledo-Hernández et al. 2023), which confirmed the results of this study that coffee husk powder had a higher moisture content compared to gamal wood powder (**Fig. 3**). This phenomenon might be due to the differences in the chemical content of the two ingredients. Coffee husk has a hemicellulose content of 33.5% (de Carvalho et al. 2018), while gamal wood has a hemicellulose content of 16.81% (La O et al. 2018). Hemicellulose is amorphous and can bind water (Chen 2014). This results in materials that have a high hemicellulose content, which will absorb water more efficiently.

Fig. 3. The moisture content of biopellets with various composition variations; different letters indicate significantly different results.

3.3. Ash Content

The analysis showed that the ash content studied ranged from 2.95%–5.48%; the highest was found in the G0C100 composition, and the lowest was found in the G100C0 composition (**Fig. 4**). Based on the analysis of variance, it shows that adding coffee husks to gamal wood biopellets has a significant effect ($p \le 0.01$) on the ash content of biopellets. In general, the ash content of biopellets does not meet the target of SNI 8021:2014, but the G100C0 variation reaches the EN 14961-2 (EN-B). Coffee husks have a higher ash content than gamal wood, so adding coffee husks can increase the ash content of the hybrid biopellets. Dadi et al. (2018) showed that the ash content of coffee husk can reach 13%. The coffee husk contains highly extractive substances, so the content of minerals such as calcium and potassium in ash is relatively high (Fernández et al. 2023). Only composition G100C0 reached the EN 14961-2 (EN-B) target, and this is thought to be due to the high ash content in the coffee husk and the particle size of the gamal wood powder. Gunawan et al. (2023) studied gamal wood biopellets with a particle size of 5 mesh gamal wood powder with an ash content of 1.2%. The more prominent the particle size, the lower the volatile matter, leaving less ash in the form of combustion products than the smaller particle size (Thoyeb et al. 2021). The ash content value in this study tends to increase due to coffee husk addition in gamal wood pellets. This phenomenon could decrease the high heating value of the biopellets (Miranda et al. 2015). The ash content directly impacted the calorific value of the biopellets in this study, indicated by decreasing the calorific value (**Fig. 7**).

Fig. 4. Ash content of biopellets with various composition variations; different letters indicate significantly different results.

3.4. Volatile Matter

The analysis showed that the volatile matter's value ranged from 68.77%–81.13% (**Fig. 5**). The analysis of variance stated that adding coffee husks into gamal wood biopellets had a significant effect ($p \le 0.01$) on the volatile matter of biopellets. Overall, only the composition of G100C0 did not reach the target of SNI 8021:2014. Adding coffee husk into gamal wood biopellets can reduce the volatile matter of gamal wood biopellets. This phenomenon may be due to the nitrogen content in coffee husks that can bind aromatic compounds (Rinawati et al. 2019), which are part of the volatile matter in wood. Tewari et al. (2013) reported that aromatic compounds that lack electrons will be bound by nitrogen. After binding to these aromatic compounds, nitrogen can bind to other aromatic compounds that also lack electrons. The value of the volatile matter of biopellets found is lower than the previous research, which mentioned the value of volatile matter of gamal wood biopellets as much as 82.96% (Rahayu et al. 2022). Decreasing volatile matter was important for biopellet manufacturing because high levels of volatile matter would cause a faster combustion process, and the fuel would run out faster, making it less desirable as a fuel (Tenorio et al. 2015).

Fig. 5. Volatile matter of biopellets with various composition variations; different letters indicate significantly different results.

3.5. Fixed Carbon

Fig. 6 shows that the value of fixed carbon studied ranged from 15.92%–26.10%, with the highest fixed carbon found in the composition of G25C75 and the lowest fixed carbon found in the composition of G100C0. The analysis of variance showed that adding coffee husks to gamal wood biopellets had a significant effect ($p \le 0.05$) on the fixed carbon of biopellets. Overall, the biopellet value of all composition variations reached the target of SNI 8021:2014, which is above 14%. The addition of coffee husk will increase fixed carbon. Fixed carbon is closely related to ash content and volatile matter. Coffee husk has a reasonably high ash content, but due to the low level of volatile matter (**Fig. 5**), the value of fixed carbon of gamal wood biopellets with a mixture of coffee husk can increase. This phenomenon follows Lubwama et al. (2022), who showed that lost volatile matter will increase fixed carbon. Fixed carbon was a property that determined the amount of solid material that could be combusted after a volatile matter had been removed from the raw material (Wibowo et al. 2021). Therefore, a decrease in the value of volatile matter (**Fig. 5**) caused the proportion of fixed carbon to be higher so that the amount of solid material that could burn became more significant.

Fig. 6. Fixed carbon of biopellets with various composition variations; different letters indicate significantly different results.

3.6. Calorific Value

The analysis showed that the calorific values studied ranged from 4,061–4,290 cal/g, with the highest calorific value found in the G100C0 composition and the lowest in the G0C100 composition (**Fig. 7**). Calorific value is a fuel property that states the fuel's energy content. The high calorific value of biopellets enabled them to yield a high amount of energy in a low fuel volume (Wibowo et al. 2021). The analysis of variance showed that adding coffee husks to gamal wood biopellets had a significant effect ($p \le 0.05$) on the calorific value of biopellets. Overall, the calorific value of biopellets has reached the target of SNI 8021:2014 and EN 14961-2. Adding coffee husk to gamal wood biopellets can reduce the calorific value of biopellets. Differences in the calorific value of different raw materials cause this result. Rusdianto (2014) reported that raw materials with high calorific value would produce biopellets with high calorific value. Fixed carbon content is very influential on the calorific value. The calorific value increases as the fixed carbon content increases (Ivanovski et al. 2023). This phenomenon causes the calorific value of coffee husks to fulfill the SNI 8021: 2014 and EN 14961-2 standards, even though coffee husks

have high moisture content and ash content. The high moisture content (**Fig. 3**) and ash content (**Fig. 4**) in gamal wood biopellets with the addition of coffee husk is the cause of the reduced calorific value of biopellets. This finding follows the research of Umar and Setiawan (2022), who found that high moisture content can reduce combustion optimization because the heat will evaporate the water first before finally burning the material in the biopellet. Hansted et al. (2018) showed that ash content could reduce the calorific value by inhibiting combustion by leaving combustion products in ash. Torefaction or pyrolysis treatment of biopellets could overcome the decrease in calorific value due to increased ash content. These treatments are proven to reduce volatile matter, increase fixed carbon, and increase energy density (Rani et al. 2022; Sulistio et al. 2020). Therefore, the treatments could be done for further research.

Fig. 7. The calorific value of biopellets with various composition variations; different letters indicate significantly different results.

3.7. FTIR Analysis

The functional groups found in all samples indicate the presence of lignocellulosic structural components, proving that the raw materials used, coffee husk and gamal wood, have the same good potential to be used as biopellets for energy. Nawawi et al. (2018) reported that chemical components can be used as a parameter whether or not a material is suitable for bioenergy. **Fig. 8**. displays several peaks in all samples, such as at wave numbers 3307 cm^{-1} , 2928 cm^{-1} , 1612 cm^{-1} , and 1022 cm-1 . The complete interpretation can be seen in **Table 2**.

Based on the interpretation that has been done, the wave peak at wave number 3307 cm^{-1} indicates the presence of structural lignocellulosic polymers such as cellulose, lignin, and hemicellulose (Fabiyi and Ogunleye 2015; Hergert 1971). The wave peak at wave number 2928 cm-1 indicates the presence of C-H stretching, which means carboxylic acids are in the tested sample (Fabiyi and Ogunleye 2015; Pandey 1999). The wave peaks at 1612 cm⁻¹ and 1022 cm⁻¹ indicate the presence of lignin in the sample (Fabiyi and Ogunleye 2015; Pandey 1999). The presence of lignin strengthens the evidence that the raw materials used are very suitable as bioenergy raw materials (Nawawi et al. 2018). The wave peaks at 1058 cm⁻¹ indicate the stretching of C-N, meaning the samples have a nitrogen component (Smith 2019). The presence of organic nitrogen compounds strengthens the presumption that the phenomenon of decreasing volatile matter content is caused by aromatic compounds bound by nitrogen in the hybrid biopellets of gamal wood and coffee husk (**Fig. 5**). The addition of coffee husk to gamal biopellets strengthens the presence of carboxylic acids and lignocellulosic polymers such as lignin. This tendency is indicated by the strengthening of the appearance of peaks at wave numbers 2928 cm⁻¹, 1612 cm⁻¹, and 1022 cm-1 (**Fig. 8**). This phenomenon proves the increase in fixed carbon value (**Fig. 6**) in hybrid biopellets from gamal wood and coffee husk.

Fig. 8. FTIR analysis results of biopellets of gamal wood and coffee husk mixture at various variations of mixture composition.

4. Conclusions

Adding coffee husk can reduce gamal wood biopellets volatile matter to fulfill the requirement of the Indonesian Standard for Wood Pellet (SNI 8021: 2014). Adding coffee husk also increased the fixed carbon and density of gamal wood biopellets. However, adding coffee husk degraded several qualities, for example, increasing the moisture and ash content of gamal wood biopellets and reducing the calorific value of gamal wood biopellets. FTIR analysis proves the phenomena in hybrid biopellets, such as decreasing volatile matter and the increasing fixed carbon value caused by adding coffee husk into the gamal wood pellet. Biopellets from gamal wood and coffee husk produced the best quality biopellets in the composition of 75% gamal wood and 25% coffee husk by achieving the target criteria of SNI 8021: 2014 and EN 14961-2, except for ash content. Torefaction or pyrolysis treatment of the hybrid biopellets was recommended for

further research and was thought to overcome the decrease in calorific value due to increased ash content.

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