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Characteristics of briquettes from plastic pyrolysis by-products

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This article contributes to: 7 AFFORDABLE AND CLEAN ENERGY — SUSTAINABLE DEVELOPMENT GOALS

Highlights:

- Seven samples of char by-products from the pyrolysis process of low-density polyethylene (LDPE) plastic at various reaction temperatures and catalyst types were studied.
- The proximate test is used to determine the properties of char such as moisture content, ash, volatile matter, and fixed carbon.
- The impact resistance index (IRI) was used to test the performance of the briquettes.
- High volatile matter content in some samples indicates imperfect devolatilization.

Abstract

Pyrolysis has been proven as a method to reduce plastic waste and produce useful products, especially liquid fuels. However, plastic pyrolysis also produces gases and char as by-products which are being investigated for useful products. Therefore, our present study aims to investigate the char characteristics of plastic pyrolysis for further use as briquettes. Seven samples of char by-products from the pyrolysis process of low-density polyethylene (LDPE) plastic at various reaction temperatures and catalyst types were studied. The proximate test is used to determine the properties of char such as moisture content, ash, volatile matter, and fixed carbon while the bomb calorimeter is used to determine the calorific value. Briquettes are formed by mixing 4 grams of char and 0.5-1 gram of binder (1% starch and 90% water). The briquettes were formed into solid cylinders with a diameter of 1.75 cm and formed with a pressure of 10 kg/cm2. Furthermore, the impact resistance index (IRI) was used to test the performance of the briquettes and showed an IRI value between 100 and 200. However, of the seven char samples tested, three of them were impossible to process into briquettes because they melted during the combustion test.

Keywords: Plastic waste; Pyrolysis; Char, Catalyst, Briquettes

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1. Introduction

Global plastic production has increased over the years due to its increasingly widespread applications in many sectors [1]. As the economy has developed, the production and consumption of plastics have increased markedly to meet demand and provide practical benefits in applications such as agriculture, electronics, automotive, construction, home furnishings, and packaging [2]. However, the massive use of plastic also creates environmental problems due to its non-biodegradable nature [3]. Therefore, efforts are being made to manage plastic waste and research to convert plastic waste into other products to find alternative solutions that are cleaner and more sustainable [4]–[6].

The trend of plastic production is expected to increase over the next few decades [7] and it results in a substantial accumulation of plastic waste, requires a large space, and increases the burden on the environment [8]. Conventional techniques, such as landfilling and burning, produce exhaust emissions (CO_2, CO, NOx, and SOx) and various other pollutant particles that are harmful to health [9]. Therefore, the application of the reuse, reduce, and recycle (3R) method is very important to minimize the negative impact of waste disposal on human health and the environment [10]. In addition, promoting awareness and education about proper waste management practices can encourage individuals to adopt sustainable behaviors [11].

Among several options for reducing plastic waste, the recycling concept is the most promising method. Plastic waste is considered a cheap and abundant raw material to be converted into other useful products [12]. Processing plastic waste into oil is an interesting way to produce new fuel while improving the quality of handling plastic waste. It can be done by pyrolysis, where a thermal process is applied to decompose molecules without oxygen [13]. This process is not only environmentally friendly but also cost-effective, making it a promising solution to the growing problem of plastic waste. However, further research is needed to optimize the process, ensure safety, and maintain sustainability [14].

Until now, plastic pyrolysis has been oriented toward the manufacture of several types of oil-based fuels. In general, by-products in the form of non-condensable gas and char have not been properly utilized. These non-condensable gases can be utilized, for example, to heat reactors or to be processed into useful compounds [15], [16]. Likewise, char which has a high carbon content can be processed into fuel besides being used as fertilizer (biochar) [17]–[19]. According to Feng [20], cracking temperature and residence time significantly affect the carbon and char content formed. Cracking temperature is the temperature reached before the plastic begins to decompose. Meanwhile, the residence time, especially in the slow pyrolysis method, is the time needed to produce the product. In addition, another important parameter is the heating rate, which is the temperature rise when the plastic is attached to a hot surface until it decomposes and evaporates [21], [22].

The type of plastic used as raw material for pyrolysis also affects the volume of char produced. A study revealed that variations in the proportion of raw materials present some interesting findings [23]. When compared to products in gaseous form, char is easier to process and store as a fuel reserve. As previously explained, several factors affect the end-product of a plastic pyrolysis process. The amount of char, gas, and liquid fuel produced can vary depending on the temperature setting of the reactor. The higher the temperature of the reactor, the more oil, gas, and char will be produced. However, if there is overtemperature, oil production decreases, and gas production increases. On the other hand, the percentage of char and solids will be higher at lower reactor operating temperatures [24], [25]. In addition, the use of a catalyst affects the character of the type of compound used. Therefore, finding the optimum temperature and the right catalyst is critical to maximizing oil production while minimizing gas production and the formation of char or solids in the reactor [26]–[28]. This requires careful experimentation and analysis of the chemical reactions involved in the process.

In our present work, the application of pyrolysis techniques is not only intended to produce more liquid fuel, but also to optimize its by-products. As is known, char is a by-product of pyrolysis which is easier to process than gas. Although the amount is relatively small, it has the potential to be further processed into an energy source. Most of the chemical content of char is solid carbon, so it is possible to process it into briquettes. Density is a parameter that determines the strength of the briquettes, where the higher the formation pressure, the higher the density. In this research, the strength test refers to the ASTM D 440-86 standard by carrying out the Impact Resistance Index (IRI) [29]. Furthermore, the proximate test and ultimate test were also carried out to identify the quality of the plastic pyrolysis residue.

2. Material and Methods

The materials used in this study were seven samples of char resulting from the pyrolysis of LDPE (low-density polyethylene) plastic waste with several different treatments during the process. Samples were distinguished by treatment with variations in the amount and type of catalyst, as shown in **Table 1**. The pyrolysis process is an effective method for converting plastic waste into valuable products. The use of different catalysts during the process can significantly affect the properties of the resulting char samples. The reactor used for the pyrolysis process is a batch reactor type with a slow pyrolysis concept, as shown in **Figure 1**. The plastic capacity is 1 kg with pyrolysis reactor temperatures ranging from 450 °C to 500 °C. The data in **Table 1** shows the different variations in plastic waste treatment samples and their corresponding results. These results can be used to optimize the pyrolysis process for maximum efficiency.

Table 1. The pyrolysis treatment was observed

No. sample	Materials	Operating Temperature (°C)	Catalyst type	Catalyst weight (gram)
1	LDPE	450	Natural zeolite	300
2	LDPE	500	Natural zeolite	100
3	LDPE	450	Natural zeolite	0
4	LDPE	450	Zeolite Y	100
5	LDPE	450	Natural zeolite	400
6	LDPE	450	Natural zeolite	100
7	LDPE	450	Natural zeolite	200

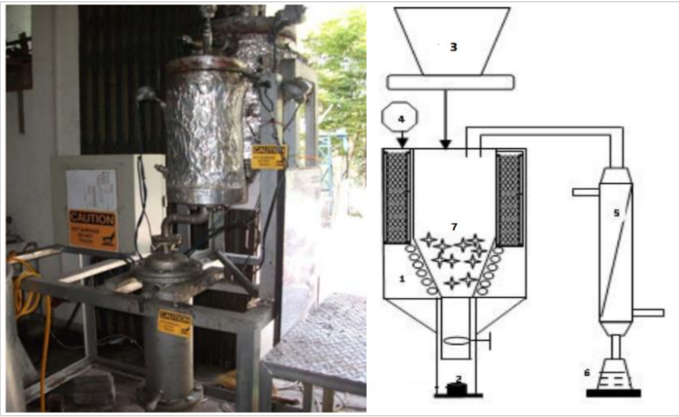

Figure 1. Pyrolysis reactor equipment: 1) Reactor; 2) Residue char; 3) Hopper; 4) Thermometer; 5) Condenser; 6) Oil pan; 7) Feedstock

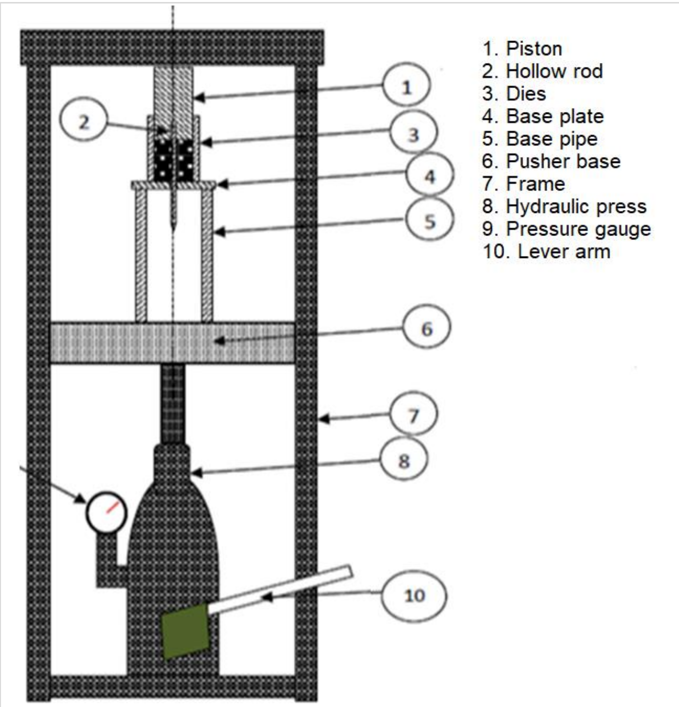

Figure 2. Unit briquette hydraulic press

The briquettes that are made are products with raw material char resulting from the pyrolysis of LDPE plastic waste without carbon admixture from other materials. Char pyrolysis results were subjected to a proximate test to determine water content, volatile matter, ash content, fixed carbon, and calorific value. The char material was made into briquettes for the test sample with a composition of 4 grams of char mixed with 0.5–1 gram of binder (1% starch and 90% water).

The briquettes were made in a cylindrical shape with a diameter of 1.75 cm and a pressing pressure of 10 kg/cm^2. The briquette pressing tool uses a hydraulic press equipped with a pressure gauge (**Figure 2**). The strength of this briquette is important because it is related to ease and durability, so that it does not break easily during transportation and use. The hydraulic press allows for consistent and precise pressure application, resulting in uniform briquette strength. This ensures that the briquettes can withstand handling and usage without disintegrating or crumbling.

3. Results and Discussion

3.1. Proximate Char Test

The characteristics of char, which is a product of the plastic pyrolysis process, can be determined through the proximate test. The data shows that each char sample has a ratio between ash content (ash), moisture, volatile, and fixed carbon content at different percentages. The identity of each sample number is presented in **Table 2**. The proximate test is a useful method to determine the composition of the char samples obtained from the plastic pyrolysis process. This information can be used to optimize the process and improve the quality of the final product.

Table 2. Proximate char pyrolysis test results for LDPE

Sample	Proximate test				
	Heating value (cal/g)	Ash (%)	Moisture (%)	Volatile (%)	Fixed carbon (%)
1	6562.85	23.57	5.81	55.46	15.15
2	3840.14	41.06	9.28	24.68	24.97
3	4229.48	36.51	4.69	6.24	52.56
4	7583.17	21.58	4.7	60.94	12.77
5	4128.47	38.35	6.02	24.72	30.91
6	7531.75	19.77	1.99	64.59	13.63
7	7169.70	22.64	5.51	60.32	11.52

The interesting differences in properties to observe are in samples made from pyrolysis LDPE with a reactor temperature of 450 °C (samples 1, 2, 4, and 5). The average volatile matter content in the char samples was still higher than the fixed carbon content, except for sample 5, which had 6.18% more fixed carbon than volatile matter due to the influence of the percentage of more zeolite. Sample char number 2, which was obtained through pyrolysis at 500 °C, has a fixed carbon content of 0.295% higher than volatile matter. These findings suggest that the pyrolysis temperature plays a crucial role in determining the fixed carbon content of the char samples. Further studies could explore the effect of other parameters, such as heating rate and residence time, on the char properties.

The large value of the volatile matter in char indicates that there is a possibility that the pyrolysis process that has been carried out is still in the incomplete devolatilization stage. Moisture content and ash content (ash) are important parts to be analyzed. The percentage of ash as a non-combustible material will greatly reduce the percentage of other useful content in char. In addition to the ash, as a raw material for briquettes, the moisture content is an important part to pay attention to. According to research, moisture has a large influence on changes in calorific value during combustion. In addition to affecting the calorific value, according to the effect of moisture on briquettes, levels of 6–8% will provide good and strong quality briquettes, while levels of 10% or more will have an impact on weakening the strength of briquettes [30]. Therefore, it is important to consider the moisture content of the briquettes during production to ensure optimal quality and performance. Additionally, the calorific value of briquettes can be further improved by incorporating additives such as binders and fillers.

The effect of reactor temperature during the pyrolysis process can be observed through the proximate test data for 100% PE with 100 g NZ (samples 2 and 6). The observed reactor temperatures were between 450 °C and 500 °C. The higher reaction temperature indicates that the percentage of volatiles will decrease, as shown in **Figure 3**a. The decrease in the volatile content, sample with reaction temperature of 450 °C had a volatile content of 64.594%, while sample with sample with reaction temperature of 500 °C had a volatile content of 24.681%. However, the condition of the data is limited to the temperature used during the study, so at higher temperatures (> 500 °C), no graphs of changes in volatile levels are obtained.

Changes in volatile levels, as seen, have a different trend from fixed carbon levels, which, on the contrary, will increase according to the determination of the higher reactor temperature. The graph shows an increase in the percentage of fixed carbon. With pyrolysis at 450 °C, the value is 13.637%; at a temperature of 500 °C, it is higher at 24.976%. Effect of pyrolysis reactor temperature on volatile and fixed carbon content. However, if you see that the moisture content has actually increased, it is necessary to review the handling and storage conditions of the char so that there may be an influence of environmental humidity that is in direct contact with the char. n addition, the pyrolysis process carried out using waste raw materials with different conditions from the waste content in the plastic, such as additives, impurities, or other compounds, although both use the same type of PE plastic, with the type of additives and uncontrolled raw material composition allowing for different proximate data.

The difference in pyrolysis temperature shows the effect that occurs on changes in the calorific value of char, where at a temperature of 500 °C, it seems to have decreased. **Figure 3**b shows the change in the calorific value, which is 7531.748 cal/g at 450 °C pyrolysis temperature, and it decreases to 3840.142 cal/g at 500 °C. The increase in carbon content reduces the calorific value of the char since carbon has a lower calorific value than the volatile components. The degree of carbonization is another element that influences the calorific value of char. At high temperatures, the char undergoes increased carbonization, resulting in a denser and more solid structure. This denser structure reduces the char's surface area, which reduces its reactivity and combustion efficiency, resulting in a decrease in calorific value. Observing the data above, what is interesting is the fixed carbon content which increases at a temperature of 500 °C while the heating value decreases. Theoretically, the calorific value will increase in proportion to the carbon content. Therefore, it should be remembered that in a proximate test, fixed carbon is a grade that does not consider the carbon content in the volatile matter.

The calculation of the heating value is using the total carbon content measured through the ultimate test. The condition of sample 6 which actually has a high calorific value is due to the large volatile content of 64.594%, while sample 2 although the fixed carbon content is higher, the volatile content is only 24.681%. In addition, the ash and moisture content of Sample 2 is much higher, causing the calorific value per unit mass to be lower than that of Sample 6. The types of char from LDPE pyrolysis with Natural Zeolite catalyst in addition to Samples 2 and 6 that have been discussed previously have characteristics based on proximate tests as data presented in **Figure 4**.

Thus, it can be assumed that the catalyst does not affect the char composition. However, from the samples obtained, different data were obtained, possibly related to the amount of raw material and the duration of pyrolysis time used. **Figure 5** shows the data for char obtained from the process without catalyst and with Y Zeolite. Samples 3 and 4 are still char obtained from LDPE pyrolysis at 450 °C. The results for char produced without a catalyst and using type Y zeolite. Samples 3 and 4 are still charred following 450 °C LDPE pyrolysis. The findings show that utilizing type Y zeolite as a catalyst during the pyrolysis process increases char yield. This suggests that the use of type Y zeolite could potentially improve the efficiency and sustainability of pyrolysis processes for producing char, which has various industrial applications, such as in the production of activated carbon.

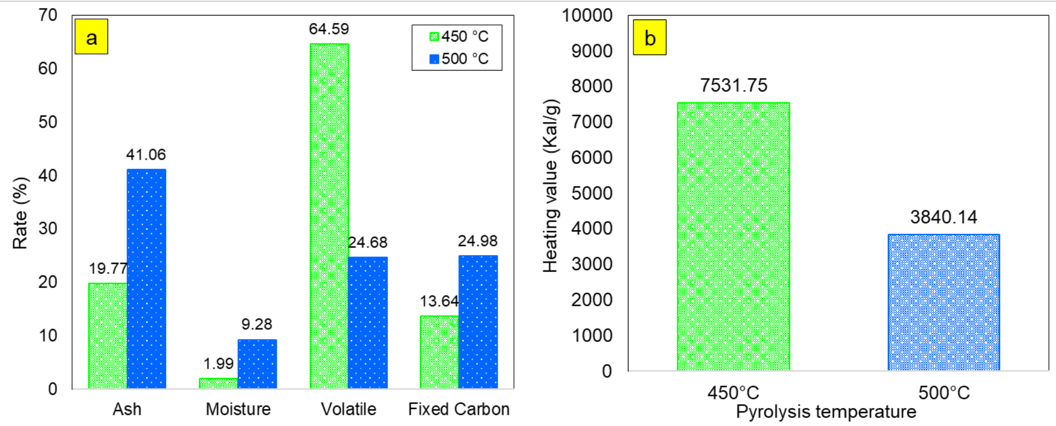

Figure 3. Effect of reactor temperature on char composition (a) and caloric value (b)

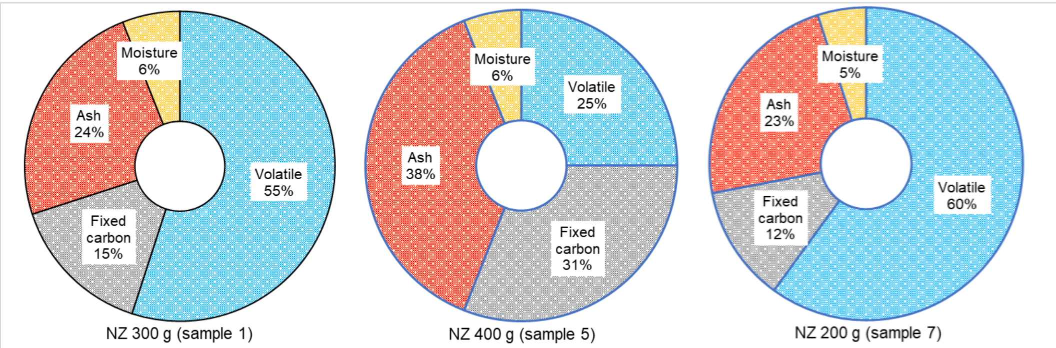

Figure 4. Proximate test char data from LDPE pyrolysis with the amount of natural zeolite catalyst

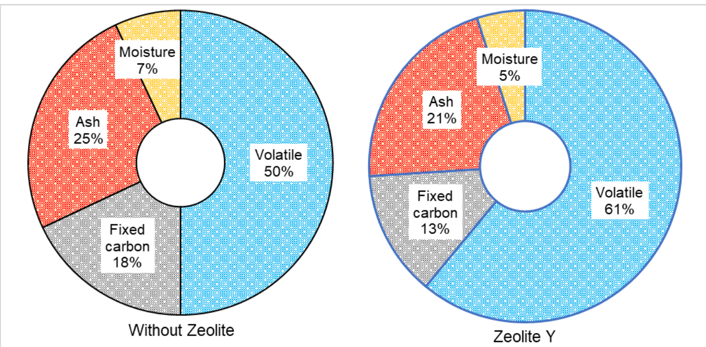

Figure 5. LDPE char proximate test data between zeolite Y and without catalyst

3.2. Characteristics of Char

The char is made into briquettes at a later stage, but due to its physical properties and characteristics, not all samples of char can be made into briquettes. The problem is that some physical properties are soft and melt when exposed to heat, as shown in **Figure 6**. Of course, this is not feasible if applied as briquettes to be burned in the furnace because the melt will merge with each briquette or even drip out of the air holes in the furnace. This is why soft char will not be tested for combustion characteristics because the melt can drip into the test equipment. These conditions will need to be improved to obtain accurate combustion data.

Figure 6. Photographic view of char during combustion

In the analysis of these physical properties, it is predicted that char can melt when exposed to heat as if it still had the properties of a raw material polymer. Observing highly volatile data indicates that devolatilization is incomplete, or in polymer pyrolysis, it can be said that depolymerization is incomplete. Based on the analysis, the char unsuitable for making briquettes are Samples 3, 6, and 7. Meanwhile, the char that can be made into briquettes are Samples 1, 2, 4, and 5.

3.3. Char Briquettes Performance

Based on the preliminary analysis, as previously explained, it has been predicted that there are three samples of char that are not suitable for making briquettes. However, to ensure the results, the three samples were still made into briquettes and tested for combustion test. The data presented in **Table 3** can show that there are three briquettes that melted when burned and dripped in the test equipment, as shown in **Figure 7**a, so only four samples could be tested for combustion. **Figure 7**b shows that dry char will make briquettes testable because until the burning is complete, there is no melt, including the ashes, which will remain on the weighing device. Char briquettes are made in a hollow cylindrical shape which aims to make it easier to place the briquettes on a platform that is connected to a digital scale. The finished and dried briquettes are then tested for strength using the concept of impact resistance index (IRI).

Testing the strength of the briquettes through the impact resistance index shows an average value of IRI 200, which means that the briquettes are not damaged and are not easily broken. In this test, the briquettes are dropped twice from a distance of 1.83 meters onto the concrete floor, and then the briquettes will remain intact or break, calculated using IRI = (100N)/n. One of the briquettes, namely Sample 1, has an index of 100 because it was broken into two parts. Even so, the strength test value of the briquettes is limited by the dimensions and mass of the samples made in this study; this is because the amount of char obtained is very limited. In making briquettes, the high pressing pressure will increase the density. A study [31] showed that briquettes made at lower pressures (30–60 MPa) were easily broken, while the properties of briquettes made at high pressures (150–250 MPa) were more consistent and denser. Usually, production at low pressure requires additional binders, for example, starch, drops, and asphalt [32].

Table 3. Briquette dimension data and impact resistance index (IRI) values

No. Sample	Proximate test					Density	Ratio (L/D)	IRI
	Outside diameter (D) (cm)	Inside diameter (cm)	Long (L) (cm)	Weight (gram)	Volume (cm^3)			
1	1.75	0.25	1.8	4.38	4.24	1.03	1.03	100
2	1.75	0.25	1.8	4.46	4.24	1.03	1.03	200
3	melted during the combustion test							
4	1.75	0.25	1.5	4.38	3.53	1.24	0.86	200
5	1.75	0.25	1.6	4.37	3.77	1.16	0.91	200
6	melted during the combustion test							
7	melted during the combustion test							

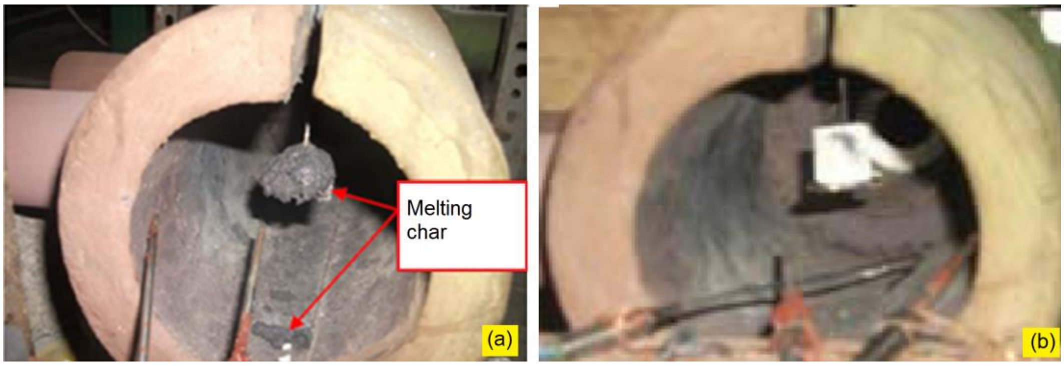

Figure 7. Melted soft char briquettes during testing (a) and char burning briquettes in dry conditions (b)

4. Conclusion

Char characteristics are strongly influenced by the pyrolysis process that has been carried out and the storage conditions after being removed from the reactor. Some samples' high volatile matter content indicates incomplete devolatilization, so if the pyrolysis time is longer, the char characteristics will differ. In addition, direct contact with the air in the storage environment results in the char having a moisture content due to the adsorption properties of carbon. As a fuel for briquettes, moisture must be removed to facilitate combustion and save energy. However, the significant carbon content with soft char particles makes the porosity of the briquettes small and inhibits the diffusion of oxygen. Further studies could explore the effect of other parameters, such as heating rate and residence time, on the char properties.

Authors' Declaration

Authors' contributions and responsibilities - The authors made substantial contributions to the conception and design of the study. The authors took responsibility for data analysis, interpretation, and discussion of results. The authors read and approved the final manuscript.

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