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Original Paper

Lignite Upgradation of Mae Moh Power Plant Thailand via Low-Temperature Pyrolysis

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***Abstract*— The lignite at Mae Moh mine in Lampang Thailand, which is the focus of this study is ranked as the lowest class in coal classification. The lignite is currently burned directly in boilers for electricity production, resulting in inefficiencies. The objective is to use low-temperature pyrolysis to improve the lignite. Lab-scale experiments were conducted using different lignite particle sizes (2.36 mm, 1.00 mm, and 0.25 mm) at temperatures ranging from 250-600°C and retention times of 0-60 mins. The best quality lignite was achieved by processing a particle size of 2.36 mm at a temperature of 350°C and a retention time of 20 mins. The gross calorific value, fixed carbon, and char content increased with higher operating temperatures. Weight loss occurred between 300-600°C due to the degradation of volatile organic fractions and char formation. Ash and sulfur constituents increased in smaller coal sizes. The average gross calorific value of char was 23.04-27.19 MJ/kg. Lower operating temperatures, larger coal sizes, and sufficient retention time are recommended for the pyrolysis process to achieve optimal results.**

Keywords*— *Pyrolysis, lignite, char, particle sizes, temperatures, retention times

I. INTRODUCTION

Coal is a rich and frequently used remaining fuel in the world. Low-quality coal is used today worldwide for energy production It is a low-cost fuel, has a high carbon to hydrogen content and is rapidly combustible. In addition to trace amounts of other components, such as minerals and water, coal also contains various amounts of carbon, hydrogen, oxygen, nitrogen, and sulfur [1]. Depending on the initial organic materials that contribute to its development, coal quality varies from location to location [2]. Natural gas, coal, hydroelectric, and oil fuels are mainly used as electricity in Thailand. The relevance of coal and coal-derived goods in supplying energy needs and supporting the country's economy will continue to grow. Energy expansion is necessary for economic growth due to recent environmental concerns and renewed interest. Thailand uses coal from many sources, both local and imported ones. The Mae Moh mine, located in the northern province of Lampang, is the most significant local source of coal in Thailand. The coal mine produces 16 million tons of lignite annually to meet the 2400 MW of overall energy requirements of the country. Mae Moh's lignite is a low-rank coal (LRC) with a lower concentration of carbon and high moisture and volatile matter. Moisture is an undesirable inherent of coal as it lowers the calorific value while volatile matter raises the risk of spontaneous ignition of lignite in a chemical process [3]. While LRC like lignite has a lower energy content compared to high-rank coals like bituminous and anthracite, it remains a valuable resource for energy production, especially in regions where other energy sources may not be readily available [4].

LRC has an extreme moisture content, which has a big impact on its utilization process, which includes dropping power plant efficiency, raising transportation costs, increasing CO_2 emissions, and causing spontaneous ignitions while being stored. LRC has a higher proportion of amorphous carbon, fewer aromatic carbon atoms and a comparatively lower aromaticity value. Coal is a carbonaceous material consisting of organic and inorganic substances [5]. LRC and lignite have low ash fusion temperatures, high sodium and/or potassium content, and a high base/acid ratio, which makes them susceptible to combustor slagging and/or fouling in high temperatures. Its poor combustion efficiency and significant impurities, along with the large amount of waste and greenhouse gas emissions, constrain the economy when it is used directly as fuel. LRC "are not conducive" to producing electricity for immediate use [6].

The efficiency of lignite coal is low, and it needs to be upgraded before use. Pyrolysis is one of the essential intermediate steps for LRC conversion technologies, including coking, combustion, gasification, and direct liquefaction [4]. The low-temperature pyrolysis upgrades the lignite by removing the moisture via elevating the operating temperature while extending the duration of residence. The pyrolysis process also enhances the increase in the gross calorific value of the coal which improves the fuel consistency, resulting in more efficient and controllable combustion [7].

Pyrolysis is a simple and inexpensive, thermochemical degradation of carbonaceous material. It is the first step before gasification and combustion in the thermochemical conversion process and occurs in the absence of oxygen. The complex process of LRC pyrolysis involves both physical and chemical changes of organic and inorganic materials, occurring at high temperatures in the absence of oxygen or with inert gas. Coal pyrolysis technology is crucial for clean and efficient use of LRC like lignite [8]. Niu et al. (2016) performed an experiment and studied the pyrolysis of lignite at the temperatures of 450, 500, 550, and 600 °C to analyze the yield distribution of product gases, char, tar, and water in N_2 and CO_2 atmosphere. At the higher temperature, the pyrolysis gases and water yield increases, while the char yield decreases. Whereas the yield of tar increases significantly to its maximum value over the temperature range of 450–500 °C, before it gets constant at temperatures above 500 °C. This is because the intensity of lignite cracking increases with increase in the pyrolysis temperature, and as char is consumed, more pyrolysis volatiles are produced [9].

Ren et al. (2019), used the microwave pyrolysis method to promote the quality of lignite. According to the findings, there was a significant reduction in oxygen-containing functional groups and an improvement in coal quality. Microwave pyrolysis effectively promoted the maximum enhancement in upgrading lignite by adding 10 wt% of petroleum coke (PC) to the feedstock. Under the structural combined synergies, the re-adsorption action of moisture was prevented, resulting in the highest decrease in moisture re-adsorption ratio (MRR) with the addition of 10 wt% PC [10].

Xu et al. (2016), developed a kinetic model for low temperature pyrolysis in a fixed bed reactor, and investigated the distribution of produce species from Zhaotong lignite. Gas chromatography (GC), Gas Chromatograph/Mass Selective Detector system (GC/MSD), Fourier transform infrared spectroscopy (FTIR), nitrogen adsorption-desorption isotherm analysis, and Scanning Electron Microscope (SEM) were used to examine the properties of the product gases. It was concluded that the temperature, heating rate and particle size play an important role in the low temperature pyrolysis. During the process, it was also suggested that atomic ratios of H/C and O/C have some correlativity with the yield of the products [11].

Qiang et al. (2021), The low-temperature pyrolysis upgrades the lignite by removing the moisture via elevating the operating temperature while extending the duration of residence. One drawback of this method is that conventional organic solvents tend to be toxic. While the pretreatment approach aims to modify the coal structure and optimize the yield of pyrolysis products, there is still a need for a more thorough understanding of the relationship between structure and activity during the thermal decomposition process [12].

Pretreatment of lignite plays a crucial task in the upgrading of LRC. Throughout the pyrolysis process for lignite, the pyrolysis tar yield is moderately low, while the yield of pyrolysis water is high. The abnormal yield of pyrolysis water is attributed to the extreme moisture content of lignite on its surface and pores. Therefore, it is economically and practically significant to develop a method that can efficiently dehydrate lignite, increase the pyrolysis tar yield, and reduce the pyrolysis synthetic water yield for the effective utilization of lignite. There are several pre-treatment mechanisms that have been studied in literature for the removal of excessive moisture in the LRC, namely thermal [13] hydrothermal [14], [15] and swelling [16] treatment processes. Liu et al. [13], studied the combined effect of hydrothermal and swelling pretreatment on LRC to upgrade the value of coal by reducing the moisture content in the as received feedstock. The outcomes demonstrated that the combined procedure was a successful way to raise lignite quality and boost pyrolysis tar yield. When hydrothermal treatment was performed at a temperature of 310 °C, the moisture content dropped to minimum levels and successfully eliminated the oxygen involving functional groups.

All the above studies focused on the upgradation of coal through different techniques and methods. The use of extra equipment to analyze the pyrolysis products incurs extra cost to the overall system. This study investigates the effects of low-temperature pyrolysis operating parameters, such as the feed particle sizes, temperatures, and retention times, on the characteristics of pyrolysis, including moisture content and volatile matter using the proximate and ultimate analysis. The quality of char from pyrolysis at low temperatures lignite coal has been investigated using weight loss and laboratory analysis. The primary focus on the laboratory and experimental results is to investigate the impact of operating parameters on several factors throughout the pyrolysis process, such as (1) the removal of moisture, (2) the production of low molecular volatiles from lignite coal, (3) the formation of char residue, (4) the increase in fixed carbon, and (5) the enhancement of gross calorific value. Char particles are usually formed from the polymerization and condensation reactions. The release of volatiles changes the macromolecule of coal, which influences the reactivity of char [17]. Analysis of ash and sulfur content in the char are also important for the upgrading process. Assessing the pyrolysis process and operating parameters is critical for improving energy utilization and environmental safety in coal technologies [18]. In this regard, the effectiveness, efficiency, and reliability of this method are evaluated for upgrading low-quality Mae Moh's lignite through the production of good quality char.

The main aim of this research is to use low-temperature pyrolysis to upgrade Mae Moh lignite and produce high-quality char as a replacement for thermal or pulverized coal. The hypothesis is that low-temperature pyrolysis can improve the quality of lignite by reducing moisture and volatile matter, thereby increasing the gross calorific value. This technique only uses the weight loss, weight loss rate and the laboratory results of proximate and ultimate analysis to examine the pyrolysis products. The pyrolysis experiments were performed on three different size fractions of coal namely, 2.36, 1.00, and 0.25 mm, to find the optimum results in terms of coal upgradation. Which is to increase the gross calorific value by decreasing the content of moisture and volatile matter. The Mae Mo power plant process line will be impacted in the future by using this research.

II. MATERIALS AND METHODS

A. Sample Preparation

The lignite samples were collected on-site from Mae Mo power plant. The power plant receives raw lignite in irregular shapes and of larger sizes. For storage purposes, the lignite size is reduced to 2.36 mm. It is further reduced to 0.25 mm for power plant usage. The original size of 2.36 mm was crushed and ground to produce a size of 1 mm in the laboratory. This process was undertaken to determine the proportionality in the research results.20 mg of lignite was used in every single experiment.

B. Experimental Setup

Lab-scale experiments were conducted using a fixed bed reactor made of stainless steel. The reactor had an internal diameter of 50 mm and a height of 62 cm. The reaction heat was provided to the reactor by an electric furnace. Electric furnace is manual set up according to the heating rate. The reactor could run at temperatures of up to 1100 °C and produce an alloy with high strength and exceptional oxidation resistance called 253 MA (253 MA obtains its heat-resistant properties by advanced control of microalloying additions), and the temperature was measured with K-type thermocouple, placed inside the reactor. Analytical balance is used for measuring the weight of samples and to calculate the weight loss of the char products. It provides the level of precision, accuracy, and sensitivity required to make accurate mass measurements. A desiccator is not used for coal pyrolysis directly but is employed in the preparation and preservation of coal samples. It helps remove moisture from coal samples before pyrolysis, ensuring accurate results, and can also be used to store pyrolyzed samples in a dry environment to prevent moisture absorption and changes in sample properties. After pyrolysis experiments, coal samples may need to be stored for further analysis or characterization. It's important to store them in a dry environment to prevent any moisture absorption that could change the properties of the samples over time. A desiccator can be used for this purpose to maintain the dryness of the samples. Nitrogen gas was supplied to the reactor at a volume flow rate of 100 ml/min, to avoid unnecessary combustion process. Nitrogen gas is used for coal pyrolysis primarily to create an inert atmosphere, exclude oxygen, control the process, maintain product quality, ensure safety, and reduce emissions of harmful pollutants. The schematic diagram of the lab – scale experimental setup is shown in Fig. 1.

In each experiment, approximately 20 grams of coal sample were initially placed into the crucible, which was subsequently inserted into the fixed-bed reactor. The samples were dried at 105°C for 20 hours to remove the moisture at a heating rate of 5°C/min. After the moisture removal, the sample was kept in the desiccator for 30 minutes. Then, it was measured with an analytical balance, and the weight loss represented the moisture content within the coal sample. The sample was operated with a higher heating rate of 10°C/min and subjected to low-temperature pyrolysis (0-600)°C for each experiment. After pyrolysis, the sample was once again placed in the desiccator for 30 minutes. The volatile content in the low-temperature pyrolysis products was measured in the coal samples, which were separated at the pyrolysis temperatures. The loss of moisture, loss of volatiles, and char residues of lignite were obtained as outputs. These outputs, along with proximate analysis data, were then used to investigate the thermal characteristics of the lignite samples.

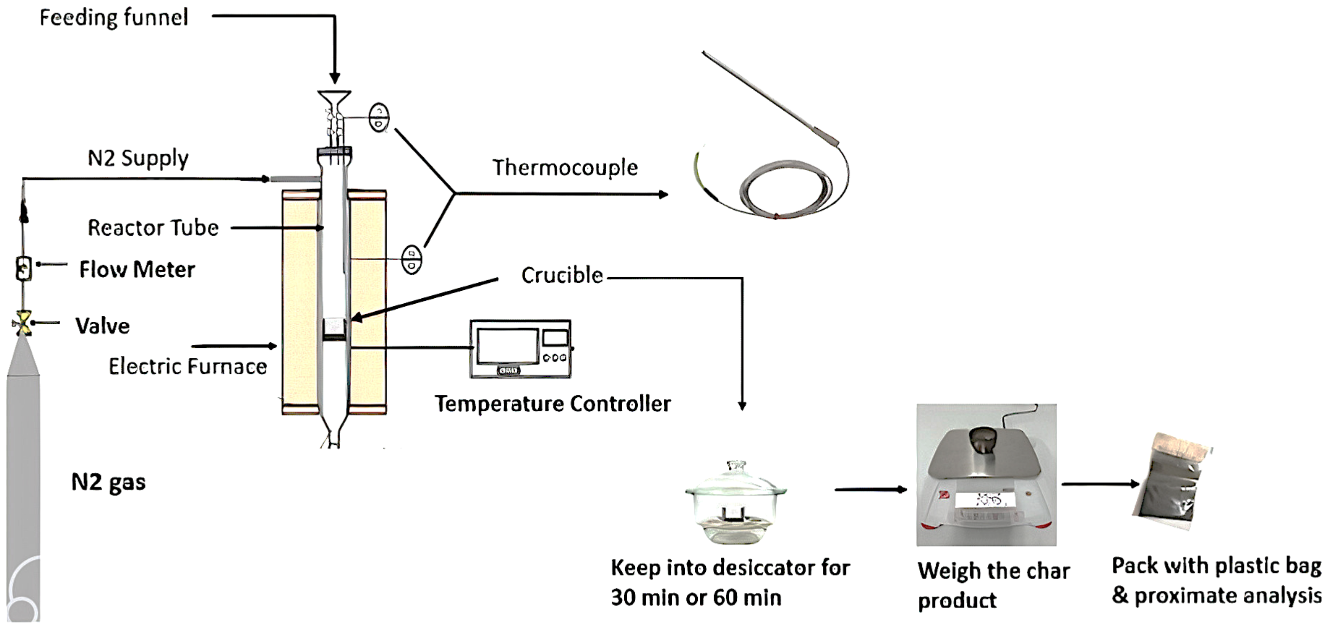
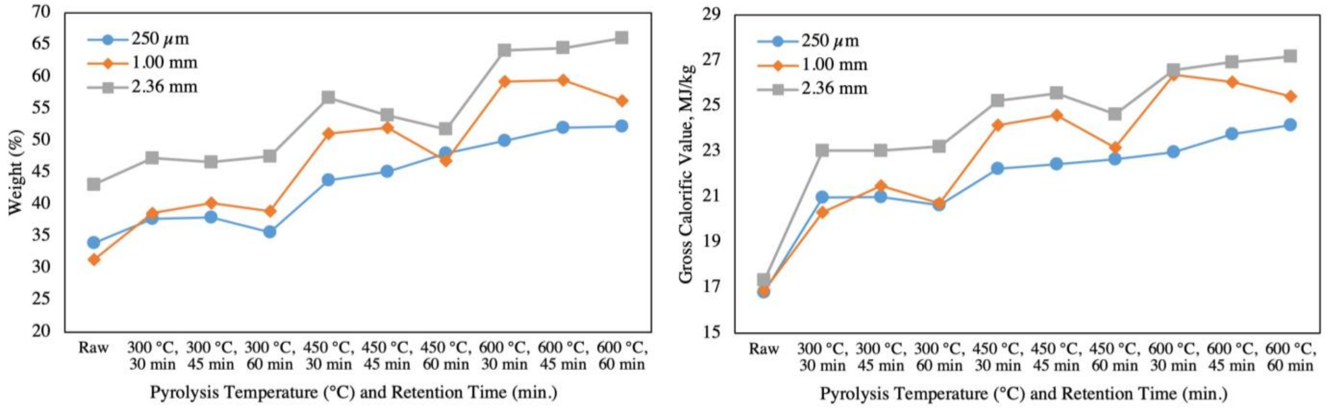


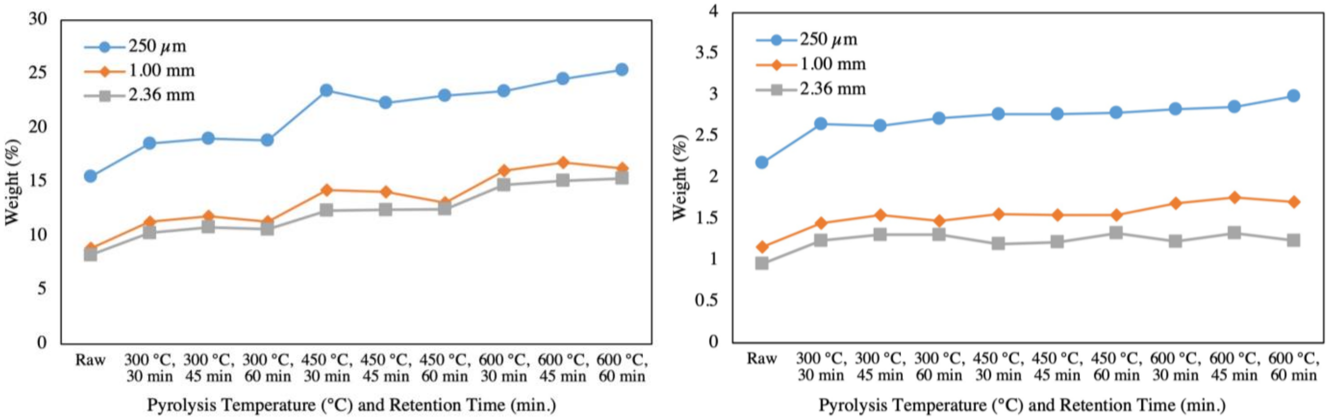
Fig. 1. Laboratory-scale experimental setup with fixed bed reactor

TABLE I. EXTENTS OF PARAMETERS IN THE INVESTIGATION.

Proximate analysis (wt%)	**Method**	**2.36 mm**	**1 mm**	**0.25 mm**
Moisture (a.r)	ASTM D3302/3302 M-17	26.75	26.61	20.20
Volatile content (d.b)	ASTM D7582-15	45.64	45.16	46.62
Ash (d.b)	ASTM D7582-15	11.25	12.10	19.42
Fixed carbon (d.b)	ASTM D7582-15	43.11	42.74	33.96
Sulfur (d.b)	ASTM D4238-19	1.32	1.57	2.73
Gross calorific value (d.b)	ASTM D5865-13	23.68 MJ/kg	23.01 MJ/kg	21.06 MJ/kg
a.r = as received d.b = dry basis				



(a) Fixed Carbon content in wt % (b) Gross Calorific Value content in wt%



(c) Ash content in wt% (d) Sulfur content in wt%

Fig. 2. Effect of particle sizes on Fixed Carbon content, Gross Calorific Value content, Ash content and Sulfur content at different pyrolysis temperatures and retention times.

C. Experimental Analysis

Lignite was pyrolyzed from room temperature (25 ˚C) to the final temperature (600 ˚C) in a nitrogen inert atmosphere. In the low-temperature pyrolysis experiment, various operating conditions were analyzed at different temperatures and retention time periods, to examine the change in properties of pyrolyzed lignite. The proximate and ultimate analysis of the raw lignite in three different sizes are shown in Table 1. The individual effects of moisture, ash, volatile matter, and fixed carbon on the calorific value of coal were further examined. The smallest size, measuring 0.25 mm, initially exhibits a 20% lower calorific value compared to the other sizes due to its lower fixed carbon content and the highest original levels of ash and sulfur. Lignite contains a significant amount of moisture, and smaller-sized particles tend to have a higher surface area-to-volume ratio. This means that smaller particles are exposed to more surface moisture, which can dilute the energy content of the lignite and reduce the GCV.

III. RESULTS AND DISCUSSION

A. *Effect of particle size*

The yields of fixed carbon, sulfur, ash, and the gross calorific value of the pyrolysis char was correlated with particle size distribution. Fig. 2(a-d) depicts the changes in proximate analysis of the pyrolyzed lignite concerning change in particle size. The quality of the char regularly decreased with an increase in the heat-treatment temperature. The results obtained revealed a moderate weight percentage of the volatile matter content of 33.43 - 17.43 wt% with fixed carbon content of 31.58% - 63.76 wt% and high calorific values of 17.34 MJ/kg - 27.19 MJ/kg for the largest size of 2.36 mm as shown in Fig. 2(a) and 2(b) respectively. When the fixed carbon content in char exceeds 40% at moderate temperatures, it is high. In Fig. 2(a), it is evident that fixed carbon has the potential to increase the char quality. In the pyrolysis process, increasing the fixed carbon content has been shown to result in higher yields of char in coal particles. There is a direct correlation between fixed carbon contended and calorific value, with higher fixed carbon resulting in higher calorific values. The lowest ash content of 8.24 - 15.33 wt% was also observed for 2.36 mm size as shown in Fig. 2(c). Ash and sulfur contents are slightly higher in temperatures above 450 °C for three sizes.

In Fig. 2a and 2b, fixed carbon refers to the carbon content in coal that remains solid or in a fixed form after the volatile matter and moisture have been driven off during pyrolysis. At higher pyrolysis temperatures, more of the volatile matter and moisture in the coal are driven off, leaving a higher proportion of fixed carbon behind. This increase in fixed carbon content typically leads to a reduction in the volatile matter content. As the temperature during coal pyrolysis increases, the GCV of the resulting char product tends to increase. This is because the higher temperatures result in the removal of volatile matter and impurities, leaving behind a more carbon-rich and energy-dense material.

In Fig 2c and 2d, the sulfur content increases as the particle size decreases. Reducing the particle size of lignite can enhance the transfer of sulfur into the gaseous phase. Xu. F et al. [17] have found that with the reducing particle size of lignite, the release amount of H_2S, COS and SO_2 increases during the pyrolysis of lignite. According to the proximate analysis and weight %, approximately 30% of the sulfur was transformed into the sulfur products and approximately 50% of ash in coal combusted at temperatures above 400 ˚C for all sizes. During coal pyrolysis, as the temperature continues to rise, the mineral matter in the coal does not decompose but instead undergoes various chemical reactions. These reactions can lead to the formation of solid ash particles. The ash particles can adhere to the surface of the char or remain as separate particles within the char matrix.

During pyrolysis, many of the bridge and branch bonds that exist in coal's molecular structure are released as volatile matter, which determines the ignition point of coal and char. LRC are a type of fuel with a high concentration of volatile matter but low fixed carbon content, resulting in low calorific values. The higher the volatile content, the lower the ignition temperature [16].

B. *Temperature effect*

Fig. 3 shows the relationship between weight loss (wt%), weight loss rate (g/min) and pyrolysis temperature. The experiments were conducted within a temperature range of 100 to 600 °C, focusing on lignite particles with a size of 2.36 mm. The results show that the weight loss rate is highest at approximately 400 °C, indicating that lignite exhibits increased reactivity and becomes more easily combustible at this temperature. The rate of weight loss in coal pyrolysis is an important parameter that describes how quickly coal decomposes or undergoes thermal degradation when subjected to high temperatures in the absence of oxygen. This indicates that they release a significant amount of volatile matter that can be readily ignited. This suggests that the lignite particles experience a more rapid decomposition, leading to a higher weight loss rate. Coal samples that have a high weight loss rate (moisture) at 250 °C and 400 °C relatively low temperatures are typically more and volatile and can ignite more easily. Volatile matter, which is a combustible component of coal, plays a significant role in this process. A higher concentration of volatile matter in lignite can contribute to more intense combustion. By increasing the volatile matter content or decreasing the coal rank (indicating a lower degree of coalification), the ignition temperature of lignite can be reduced, allowing it to ignite within a temperature range of 350 °C to 400 °C.

Temperature and residence time are crucial variables that impact the behavior and product outcomes of the pyrolysis process. The pyrolysis temperature directly influences the degree of decomposition and the formation of various products. Residence time refers to the duration that the lignite particles spend at a given temperature during pyrolysis, affecting the extent of the chemical reactions and transformations taking place. Considering the proximate analysis data, weight loss rate analysis, ignition temperature point, which provide information about the composition of coal, a pyrolysis temperature of 350 ℃ is found to be appropriate.

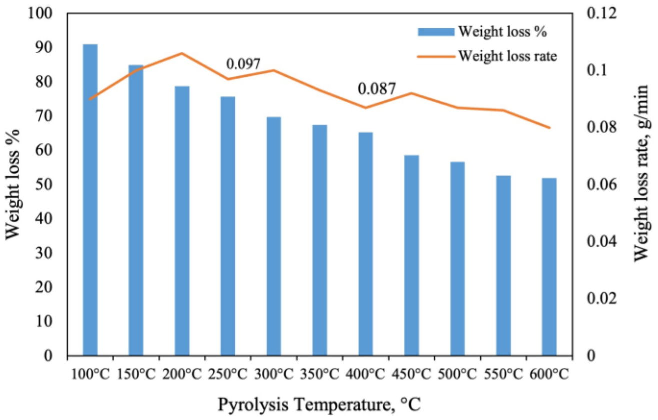


Fig. 3. Weight loss of the largest size (2.36 mm) of lignite at different operating temperatures.

C. *Retention time effect*

Fig. 4 illustrates the connection between retention times, final temperatures, and char and volatile yields in the pyrolysis of Mae Moh lignite. The data suggests that extending the operation time by 20 minutes increases char yields, signifying more extensive lignite conversion into solid char. Higher pyrolysis temperatures and longer retention times also enhance feedstock conversion but reduce char yields due to increased conversion into volatile components. Additionally, longer volatile residence times in the reactor decrease char yield, indicating further decomposition of volatiles into gases or other products. Optimizing pyrolysis conditions, like adjusting retention time and temperature, can control char and volatile product distribution, impacting pyrolysis process efficiency and output.

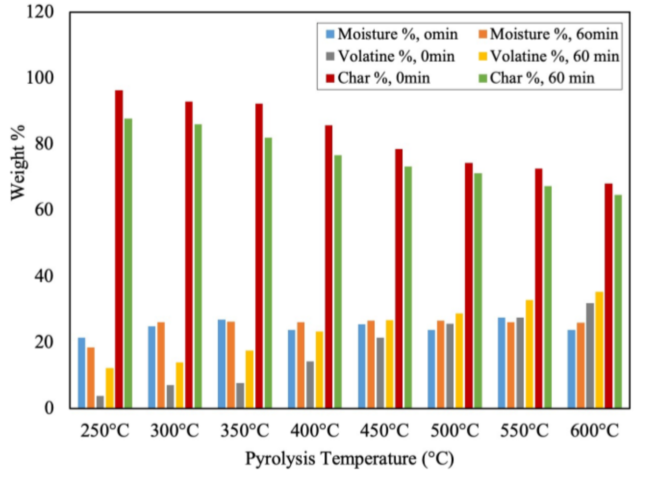


Fig. 4. Composition of moisture, volatile, and char at 0- and 60-min retention time by increasing temperatures.

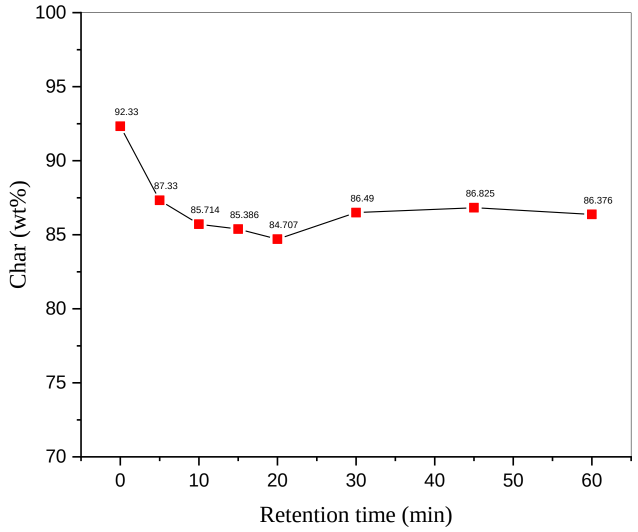


Fig. 5. Char (wt%) at optimum 2.36 mm at operating temperature of 350 with retention time of 0-60 min.

Fig. 5 illustrates that, at a temperature of 350°C and retention times between 15 and 20 minutes, the mass of the samples remains constant. When extending the retention time beyond this range, more char forms, resulting in a higher carbon content in the final product. However, there is an optimal retention time that strikes a balance between char production and achieving a high calorific value, typically falling between 10 and 30 minutes. The graph also indicates that as the pyrolysis temperature increases, the char yield decreases. This is primarily due to the thermal decomposition of polycyclic aromatic hydrocarbons (PAHs) present in the lignite. PAHs are complex organic compounds that break down at higher temperatures, leading to reduced char production [4]

IV. CONCLUSION

The heating value of coal, also known as its calorific value, depends on various factors, including its composition and the specific type of coal. In every single experiment, the largest 2.36 mm size with higher carbon content and lower moisture, ash, and impurities have a higher heating value. At a temperature of 350 ˚C and retention times of 15-20 minutes, the mass of the samples remains constant. Increasing the pyrolysis temperature leads to a decrease in char yield due to the thermal decomposition of polycyclic aromatic hydrocarbons (PAHs) present in the lignite. Longer retention times result in increased char formation and higher carbon content in the final product, but there is an optimal retention time (typically 10-30 minutes) to balance char production and calorific value.

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