

Influence of microwave pre-treated Palm Kernel Shell and Mukah Balingian coal on co-gasification

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ABSTRACT

In this study, microwave irradiation pretreatment of palm kernel shell (PKS) and Mukah Balingian (MB) coal was carried out in a fixed bed reactor. The effect of microwave power and processing time was investigated on pretreated PKS and MB coal characteristic. Then, the co-gasification of microwave pretreated PKS and MB coal was conducted to examine the effect of product yield and gases composition. The results showed that, the characteristics of pretreated sample was improved with increasing microwave power and processing time. The volatile matter, oxygen content and O/C ratio of pretreated sample decreased, while the calorific value, fixed carbon and carbon content of pretreated sample increased with increasing microwave power. The carbon content of pretreated PKS was closed to the untreated MB coal with comparable calorific value was obtained. The microwave power of 450 W and processing time of 8 min were appropriate to upgrade the PKS and MB coal for co-gasification. The pretreated sample produced higher gas yield and lower tar and char yield than the untreated sample during co-gasification. This result was due to low moisture and oxygenated compound of pretreated feedstock made it appropriate to be converted in co-gasification. Moreover, co-gasification of pretreated sample produced the higher H₂+CO and CH₄ and lower CO₂ composition than untreated sample. Thus, it can be concluded that the microwave irradiation pretreatment on PKS and MB coal performed the significant impact on the product distribution and composition during the co-gasification.

Keywords: Microwave irradiation; torrefaction; pretreatment; Palm Kernel Shell; Mukah Balingian coal; co-gasification.

INTRODUCTION

In recent years, the increasing emission of CO₂, SO_x and NO_x has become a concern on the utilization of the world energy [1, 2]. In the midst of limited availability of fossil fuels and high level of air pollution, energy efficient technologies are gaining importance and

gasification being highly efficient technology, has received significant attention [3, 4]. Currently, coal is the main feedstock in gasification and is expected to be applied as the energy resource for many decades ahead. However, this direction difficult to achieve due to the increasing in energy demand that had caused the shortage supply and reducing of high rank coal [5]. Consequently, one of the approaches is to utilise the abundant low rank coal and biomass in thermal conversion [6].

High-rank coals, comprising bituminous and anthracite coals, contain high carbon and low moisture content than low rank coals. Low rank coals include brown coal, lignite and sub-bituminous coals have lower energy content due to low carbon content. The low rank coals is almost partial of the world's entire coal deposits compared to the high rank coal [7]. The usage of low rank coals in thermal conversion is economical, due to its low pricing. However, low rank coal as a substitute to the high rank coal, has several limitations i.e. low calorific value and high moisture and oxygen content [7]. These weaknesses can be minimized by pretreatment of low rank coal [8]. Similarly, the utilization of biomass which is a renewable and environmental friendly resource during gasification imposed several problems. Untreated biomass has relatively low energy, high moisture and oxygenated compound, hygroscopic behavior and poor grindability [6-8]. Therefore, the pretreated biomass which has been improved in energy density, hydrophobicity and grindability overcome the weakness of untreated biomass, then driven to be applied in thermochemical conversion [12].

Most biomass torrefaction and preheated coal applied conventional electric heater, while there is an alternative technology designated microwave irradiation (MI). The pretreatment using MI is an effective technique for upgrading the biomass and low rank coal [13]. Unlike electrical heating methods in which heat gradually enters into samples through normal heat transfer mechanisms (convection, conduction, and radiation) [14], MI utilizes electromagnetic energy to generate heat which can penetrate deep into samples, allowing heating to initiate volumetrically [15].

Co-gasification has been investigated by numerous researchers. Krerkkaiwan et al. [16] found the synergistic effect in terms of higher gas yield, lower tar and char yield at 50 % biomass blending ratio with coal. Howaniec and Smolinski [17] reported that the co-gasification increased total gas yield and H₂ yield compared to individual gasification. Yuan et al., [18] stated that there was synergistic effects in the decreasing of char yield and increase of gas yield in co-conversion of coal/biomass mixtures. Consequently, the synergistic between biomass and coal co-gasification increases the gas yield, gasification efficiency and reactivity of char whereas reduces the tar yield. Most of the blending in co-gasification utilized untreated biomass and coal.

Dudynski et al. [19] reported that effective and stable gasification with lower tar production was obtained from torrefied pellets in comparison to untreated biomass and suggested that pretreated biomass more appropriate to be applied for co-gasification. Kuo et al. [20] produced higher syngas yield using torrefied bamboo in fixed bed reactor system. Definitely, torrefaction creates the gasification behaviour of biomass approach to that of coal where the H₂ composition in syngas of torrefied biomass comparable is with coal. Therefore, the pretreated feedstock which had been improved in their properties enhance the gasification performance and H₂ production in syngas [21]. Thus, the co-gasification of pretreated PKS and pretreated MB coal is absolutely innovative in this area.

The objective of the research was to inspect the microwave pretreatment on PKS and MB at various microwave power and processing time. In addition, the influence of microwave pretreatment on the properties of pretreated materials were determined. Co-gasification of microwave pretreated PKS and MB coal were investigated on product distribution and gases composition.

METHODS AND MATERIALS

Materials

MB coal from Sarawak, Malaysia was used as the low rank coal sample in this study. The untreated MB coal sample was pulverized and sieved through progressively finer screen to obtain particle sizes of less than 212 μm . PKS as a biomass sample was obtained from United Oil Palm Mill Sdn. Bhd., Nibong Tebal, Penang, Malaysia. PKS sample was crushed and sieved through progressively finer screen to obtain particle sizes of less than 212 μm . The untreated MB coal and PKS samples were dried in an oven at 105 °C for 24 h for rendering moisture free and finally stored in an air-tight container until the experiments and analyses were carried out.

Microwave Pretreatment

The pretreatment was carried out in a domestic microwave with technical specifications of ~240 V, 50 Hz and maximum power of 800 W. The microwave output power of 200, 300, 450 and 600 W were used in this study. During experiment each sample was put in quartz container on the rotating glass plate and place at the center of microwave. The sample was irradiated for 4, 8 and 12 min at the respective microwave power. The final temperature of the sample during the pretreatment process was measured using infrared thermometer immediately after the pretreatment process. The final weight of pretreated sample was measured once it reached the room temperature. The experiment under all of the studied parameters were performed in triplicate to ensure the measurement value and repeatability of the achieved results.

Sample Analyses

The proximate analysis which analysed the moisture, volatile matter, ash and fixed carbon content was carried out using a Mettler Toledo thermogravimetric analyser (TGA). The ultimate analysis of the samples was carried using elemental analyser CHNS-O Flash 2000. The Leco AC-350 bomb calorimeter was used to determine the calorific value (CV).

Co-gasification Experiment

The co-gasification of PKS and MB coal were carried out using fixed bed reactor with an internal diameter of 60 mm and 300 mm in height at an ambient pressure. An electric furnace surrounding the reactor was use to heat the reactor. A schematic diagram of the experimental set-up is shown in Figure 1. Approximately 5 g of sample was weighed and positioned inside the reactor. The reactor was flushed with nitrogen gas for 10 min before the experiment. Then, the samples were heated to the desired gasification temperature with heating rate of 50°C/min. A nitrogen flow rate of 0.5 L/min was continued to create an inert atmosphere inside the reactor. After the reactor reached the preferred gasification temperature, the steam

that was produced from the steam generator was introduced into the reactor, and the nitrogen flow was stopped. The steam gasification of the sample was held for 60 min. The experiment was conducted under optimise condition at gasification temperature of 767°C, biomass blending ratio of 52% and steam flow rate of 55 ml/min from our previous studies [22]

The volatile product which left the reactor from the upper side were condensed in a tar trap. The gas was collected in a gas bag every 15 min from the beginning of steam gasification. The remained solid, which is defined as char was weighted. The tar yield in the tar trap was weighted. The gas yield was calculated by difference based on the total mass balances considering the tar and char yield. The gases composition was analysed using portable gas analyser GA5000. The content of CH₄, CO₂, O₂ and balance gas was measured by gas analyser. The balance gas was assumed as H₂+CO since other gases such as C₂H₄, C₂H₆ and C₃H₈ exist in lower concentrations [23].

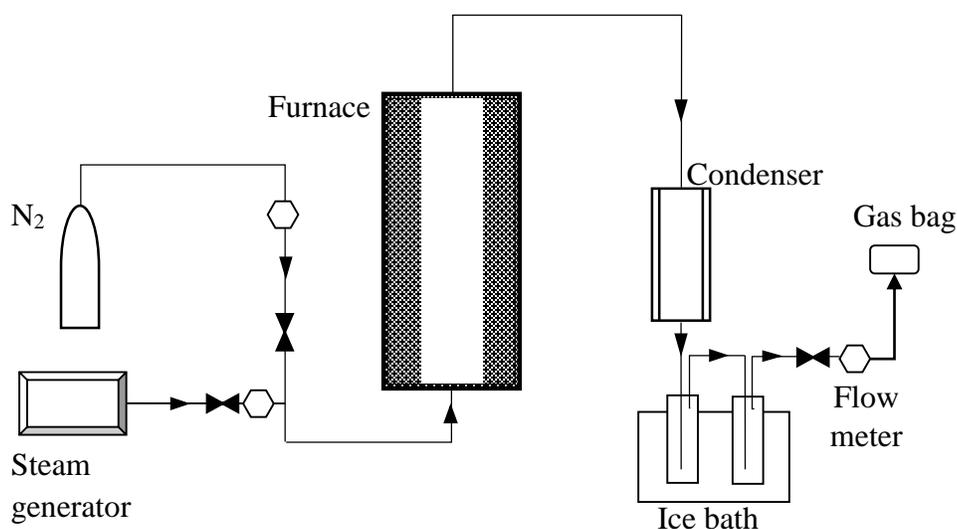


Figure 1. Schematic diagram of fixed bed reactor.

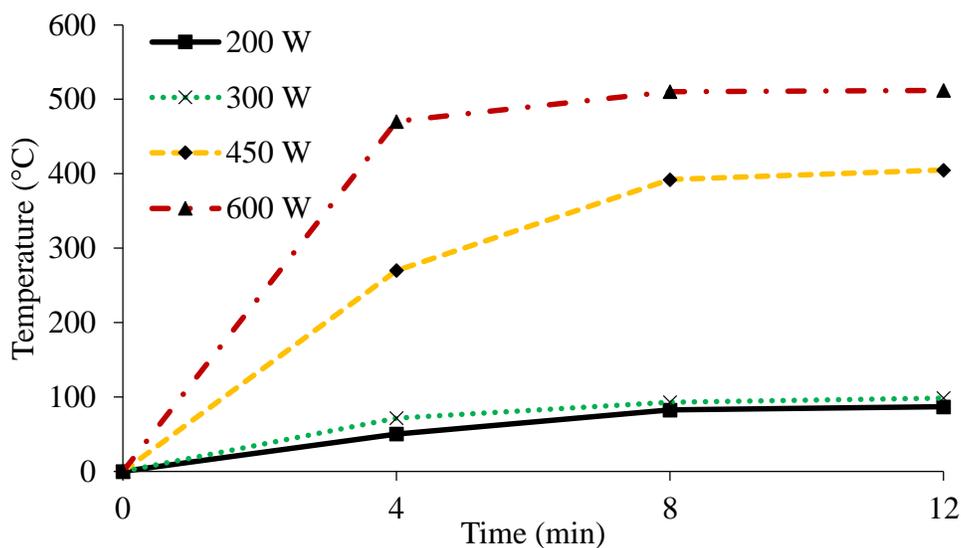
RESULTS AND DISCUSSION

Temperature Profiles of Pretreated PKS and MB Coal

Process temperature is one of the most important factors affecting thermal pretreatment. Figure 2(a) and (b) show the temperature profiles of pretreated PKS and MB coal, respectively. It shows that higher microwave power level contributed to high heating rate and final temperature. The pretreated PKS demonstrate increasing final temperature and heating rate of 50.2°C to 470.4°C and 12.6°C/min to 117.6°C/min, respectively when microwave power level increased from 200 to 600 W in the first 4 min. These temperature profiles increased much steadily about 4 to 8 min processing time. Conversely, the temperature increment is not significant at 12 min processing time regardless the microwave power. Thus, higher processing time of 12 min was not necessary to upgrade the PKS. The microwave power at 600 W with 4 min, processing time was not suitable for upgrading the PKS, where

it reached high heating rate of $117.6^{\circ}\text{C}/\text{min}$ as the torrefaction require heating rate equal or below $50^{\circ}\text{C}/\text{min}$ [24].

Figure 2(b) shows the pretreated MB coal temperature as a function of processing time for microwave power of 200, 300, 450 and 600 W. For the sample at 200 and 300 W, the sample temperature increased rapidly during the first 4 min and then the rate of temperature rise became slower and then began to level off. The sample at 450 and 600 W behaved in a similar manner but the temperature increased rapidly until 8 min processing with temperature about 110°C and subsequently followed by constant temperature stage at around 110°C until 12 min processing time. The dewatering process become more intense, rapid, and concentrated at approximately above 100°C . This temperature remained constant until most of the moisture in coal was removed. The constant temperature stage corresponds to the constant drying rate period in which the free surface moisture is removed. Then, the temperature further increased slowly to remove the remaining moisture, because more energy was required to remove water molecules that were tightly bound. In these experiments, a maximum temperature of about 116°C was observed after microwave irradiation for 12 min with 600W power. The processing time of 8 min is adequate to upgrade the MB coal. The temperature variation of pretreated MB coal during microwave drying showed rather similar trend with Indonesian low-rank coal [25] and Chinese low-rank coal [26].



(a)

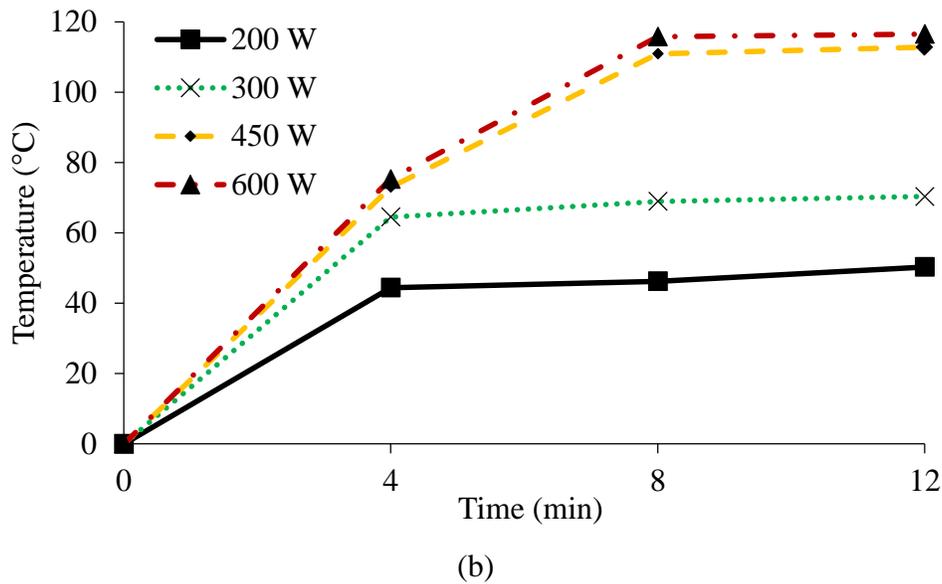


Figure 2. (a) Temperature profile of pretreated PKS, (b) temperature profile of pretreated MB coal.

Proximate Analysis of Pretreated PKS and MB Coal

The pretreated PKS and MB coal at 8 min processing time were chosen for proximate analysis based on previous section. Table 1 shows the influence of microwave power on proximate analysis and calorific value for pretreated PKS and MB coal. It can be seen that the moisture content and volatile matter decreased with increasing microwave power. Therefore, the pretreated samples properties were improved due to high moisture content of untreated sample and its capacity in absorbing microwave radiation. The fixed carbon of the pretreated PKS and MB coal increased, with increasing microwave power. The increasing of fixed carbon of the pretreated samples demonstrating an alteration in amount of energy per unit mass which associated to the calorific value. Furthermore, volatile matter decreased with increasing the microwave power due to drying, volatilization, and decomposition of biomass feedstock through the pretreatment of PKS at higher microwave power. While, through the pretreatment of MB coal, the moisture and volatile matter were released with increased in the coal reactivity [27].

The pretreated PKS had the highest CV of 20.5 MJ/kg which was 12.6 % higher than untreated PKS at microwave power of 450W. The pretreated MB coal had the highest CV of 24.7 MJ/kg which was 22.9% higher than untreated MB coal at microwave power of 600 W. Generally, higher microwave power level contributed to higher CV of pretreated feedstock.

Table 1. Influence of microwave power on proximate analysis and calorific value of pretreated PKS and MB coal.

Sample	Microwave power (W)	Proximate analysis (wt. %)				CV (MJ/kg)
		Moisture	Volatile matter	Fixed carbon	Ash	
Untreated PKS	-	10.60	77.54	10.95	0.91	18.2
PKS	200	6.81	62.28	29.99	0.92	17.8
	300	4.25	53.04	41.63	1.08	18.8
	450	1.43	46.10	50.74	1.73	20.5
	600	3.68	24.36	69.54	2.42	20.1
	Untreated MB	-	21.50	38.00	35.60	4.90
MB	200	10.39	31.45	52.96	5.20	21.5
	300	8.06	29.40	57.44	5.10	22.3
	450	6.81	29.02	58.87	5.30	23.6
	600	4.30	29.81	60.88	5.01	24.7

Carbon and Oxygen Composition and O/C Ratio of Pretreated PKS and MB Coal

Figure 3a, b, and c show the influence of microwave power on carbon and oxygen composition of pretreated PKS, pretreated MB coal and O/C ratio, respectively at 8 min processing time. Generally, the results designate that oxygen decreased while, carbon increased with the increasing microwave power. The oxygen was reduced up to 43 and 33% for pretreated PKS and MB coal, respectively. Whereas, carbon was increased up to 52 and 62% for pretreated PKS and MB coal, respectively at the highest microwave power of 600W. The decreased in oxygen composition, were mostly attributed to the destruction of hydroxyl group (-OH) in PKS and MB coal during pretreatment, which accordingly formed solid hydrophobic fuel. Eventually, by removing oxygen using MI method, the CV and energy density of the pretreated PKS and MB coal increased.

Due to the decomposition and elimination of volatile matter during pretreatment process, the oxygen composition of the pretreated products will be lowered. Therefore, as demonstrated in Figure 3(c), the O/C ratio of all pretreated samples were lower than that of the untreated sample. As the microwave power level increased, the O/C ratio of pretreated PKS and MB is gradually reduced as more volatile matter being released as a result of the continuous decomposition process. Moreover, the decrease in O/C ratio with the increasing microwave power level from 200 to 600W suggests that the coal rank has been upgraded. The reduction of the atomic ratios also indicates the measures of conversion efficiency and oxidation degree of pretreated products [25].

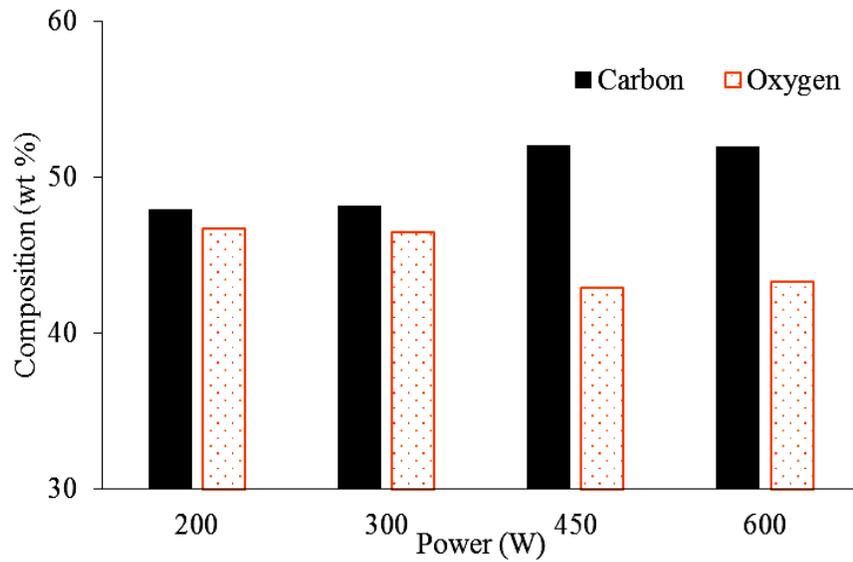


Figure 3(a). Influence of microwave power on carbon and oxygen composition of pretreated PKS.

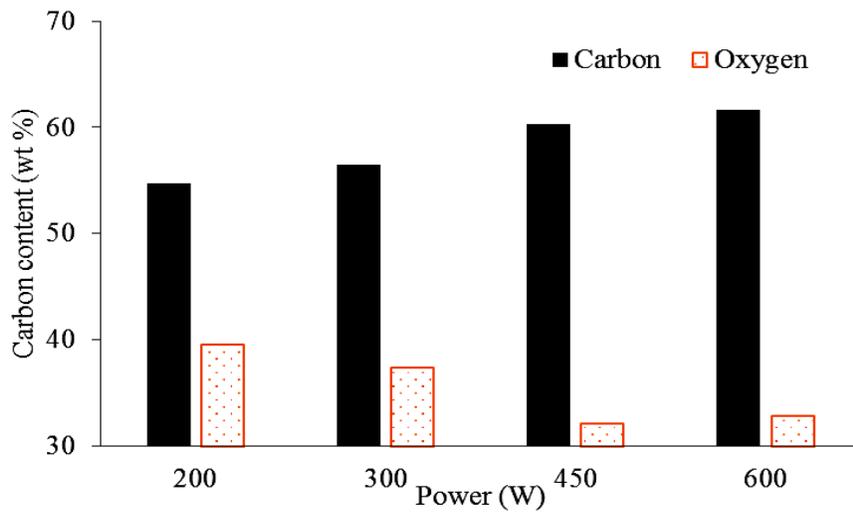


Figure 3(b). Influence of microwave power on carbon and oxygen composition of pretreated MB coal.

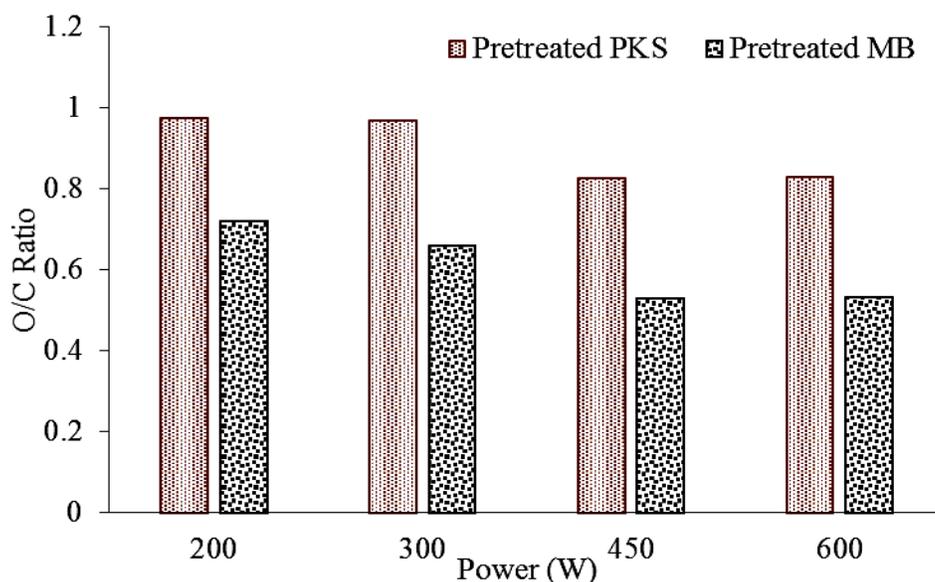


Figure 3(c). Influence of microwave power on O/C ratio of pretreated PKS and MB coal.

Co-gasification of Pretreated PKS and MB Coal

Figure 4 shows the co-gasification product yield between untreated and pretreated PKS and MB coal. The pretreated samples produced higher gas yield than the untreated sample. The gas yield increased about 20% using pretreated sample. The pretreated sample exhibited prominent impact on gas production to produce high gas yield compared to untreated sample. The gas production using pretreated sample was higher than Berruoco [26] which increase only 7% than untreated sample on gasification of pretreated Norwegian forest residue.

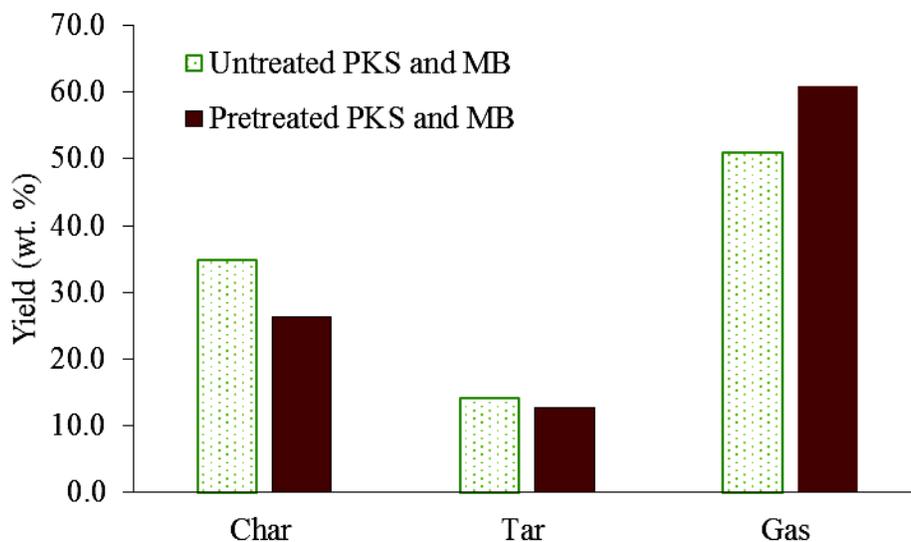


Figure 4. Effect on product yield between co-gasification of untreated and pretreated sample.

The tar yield reduces from 14.2% to 12.8% for pretreated sample. Low tar yield is required in co-gasification. Thus, the pretreated sample reduced the tar yield in co-gasification as an implication of the partial removal of oxygen composition of PKS and MB coal and volatiles through the pretreatment process. The char yield decreased about 30.2% for pretreated sample. Low char yield for pretreated sample was associated with the increased of conversion to gas product using pretreated feedstock. Moreover, this occurrence was affected from the low moisture and oxygenated compound of pretreated feedstock made it soothing to be converted in co-gasification.

Figure 5 shows the overall gases composition between co-gasification of untreated and pretreated PKS and MB coal. The H_2+CO composition using pretreated sample was higher than untreated sample. The co-gasification of pretreated sample increased the H_2+CO composition by 19.4%. These indicated that the chemical reaction such as water-gas reaction and steam reforming reaction which favour production of H_2+CO was greater on co-gasification of pretreated feedstock. The CH_4 composition showed that the co-gasification of pretreated PKS and MB coal enhanced the CH_4 than untreated sample. This proved that the co-gasification of pretreated sample able to convert the heavy hydrocarbon in tar yield to light hydrocarbon gas such as CH_4 . Co-gasification of pretreated sample exhibited reduction of CO_2 composition by 33.8 % as compared to co-gasification of untreated sample.

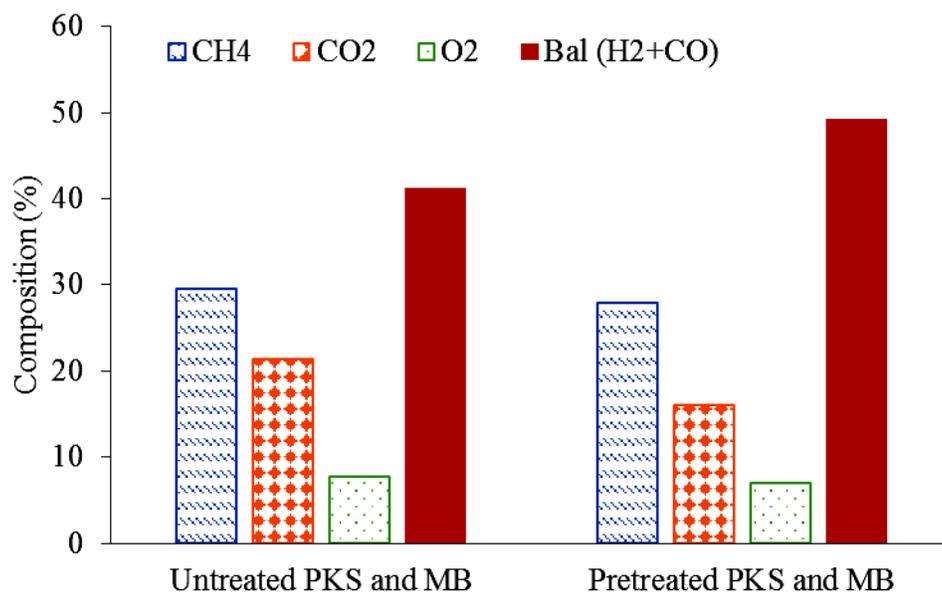


Figure 5. Effect on gases composition between co-gasification of untreated and pretreated sample.

CONCLUSIONS

The influences of microwave irradiation pretreatment on PKS and MB coal was investigated. It was determined that the CV, fixed carbon and carbon content increased, however, volatile matter and oxygen content reduced, as the microwave power increased. The pretreatment

showed the optimum parameter at microwave power of 450W and processing time of 8 min to produce a good quality fuel. The co-gasification of microwave pretreated PKS and MB coal shows a positive effect in terms of product yield distribution by producing 20 % higher gas yield than co-gasification of untreated sample. The H₂+CO composition increased by 19.4% and CO₂ composition decreased by 33.8% through co-gasification of pretreated PKS and MB coal. Consequently, the pretreated PKS and MB coal which had been enriched in their properties improved the gasification performance in term of product yield and gases composition.

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