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The Effect of Pretreated Palm Kernel Shell and Mukah Balingian Coal Co-gasification on Product Yield and Gaseous Composition

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Abstract. In this study, co-gasification of palm kernel shell (PKS) and low-rank Malaysian coal (MB) was carried out in a fixed bed reactor. For the pretreated samples, PKS was torrefied at 270°C (PKS_{To}) and MB was preheated at 250°C (MB_{Pr}) for 1 h, respectively, prior to co-gasification at 767°C, with a biomass blending ratio of 52% and a steam flow rate of 55 mL/min. The effect of different blending combinations was investigated towards product yields, namely gas, tar, char and gases composition. The co-gasification on both pretreated (PKS_{To}/MB_{Pr}) and catalyst-pretreated (Cat-PKS_{To}/MB_{Pr}) produced a greater gas yield, with lesser tar and char yield than both untreated PKS and MB (PKS_{Un}/MB_{Un}) and pretreated PKS and untreated MB (PKS_{To}/MB_{Un}). The PKS_{To}/MB_{Pr} was found to enhance the H_2 production by 63.9% and 41% than PKS_{Un}/MB_{Un} and PKS_{To}/MB_{Un}, respectively, at 45 min of reaction time. Thus, the pretreatment on both samples had a significant impact on the distribution and composition of product yields during co-gasification. As a conclusion, the pretreated sample, which has been upgraded on characteristics such as higher carbon and lower oxygen content than the untreated sample was revealed to enhance gas yield and H_2 production during co-gasification.

Keywords: Biomass; Gasification; Low rank coal; Palm kernel shell; Pretreatment

1. Introduction

Currently, the application of the world energy, which releases carbon dioxide, sulfur oxide and nitrogen oxide, has become an issue (Taba et al., 2012). The other problems are associated with the usage of fossil fuels and production of greenhouse gas. Thus, gasification, which is established as an energy-efficient technology, has an acknowledged important consideration (Sulaiman et al., 2012; Heidenreich and Foscolo, 2015). Presently, coal is the main feedstock in gasification and is expected to be applied as the energy resource for many decades ahead. However, this direction is difficult to achieve due to the increase in energy demand, which has caused the shortage of supply and the reduction of high-rank coal (Mohr et al., 2015). Consequently, one of the approaches is to utilize the abundant low-rank coal and biomass in gasification.

The low-rank coal is almost partial towards the world's entire coal deposits compared to the high-rank coal. The usage of low-rank coal in thermal conversion is economical due

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to its low pricing. However, low-rank coal as a substitute for high-rank coal has several limitations, such as low calorific value and high moisture and oxygen content (Rao et al., 2015). These drawbacks can be minimized by using the pretreated or upgraded low-rank coal in gasification (Xia et al., 2015). Similarly, the utilization of biomass, which is a renewable and environmentally friendly resource during gasification, created several problems. Untreated biomass has relatively low energy, high moisture and oxygenated compound, hygroscopic behavior and poor grindability (Chen et al., 2015). Accordingly, the pretreated biomass improved in energy density; hygroscopic characteristics and grindability overcome the disadvantage of untreated biomass and are suitable for further thermochemical conversion (Nhuchhen et al., 2014; Yuliansyah et al., 2019).

Biomass commonly has higher hydrogen content than coal and it is appropriate to mix both together. Further, the alkali and alkaline earth metals (AAEM) in biomass catalyze the gasification of char resulting from coal pyrolysis. Equally, the high silica (SiO_2) content in coal acts as an effective catalyst for tar cracking to light hydrocarbon in thermal conversion (Mallick et al., 2017). However, the gasification of biomass indicated more drawback than coal gasification, where biomass has high oxygenated compound and moisture content and low energy density (Ahmad et al., 2014; Kasim et al., 2019; Ahmad et al., 2019). Thus, co-gasification of biomass and coal can be substituted for individual gasification, as it may improve their disadvantages on both feedstocks (Brar et al., 2012).

Co-gasification has been studied by some researchers. It improved the overall gas and hydrogen composition more than individual gasification (Howaniec and Smoliński, 2013) and showed the synergistic influence in terms of high gas yield, low tar and char yield at 1:1 biomass–coal ratio (Krerkkaiwan et al., 2013). There were synergistic effects in the decrease of char yield and increase of gas yield in the co-conversion of coal-biomass blending (Yuan et al., 2012). Consequently, the synergy between biomass and coal co-gasification increases the gas yield, gasification efficiency and reactivity of char and reduces the tar yield (Winaya et al., 2015). Upgraded biomass, such as torrefied pellets, was suitable to obtain low tar yield (Dudyński et al., 2015), and torrefied bamboo was also established to produce high syngas yields (Kuo et al., 2014). Moreover, the blending of pretreated biomass and sub-bituminous coal in co-gasification was found to minimize the formation of agglomerates in fluidized bed reactors (Strege et al., 2011). Definitely, torrefaction creates the gasification behavior of the biomass in its approach to coal where the H_2 composition in the syngas of torrefied biomass is comparable with coal.

Furthermore, one of the most efficient techniques of producing higher gas qualities is steam gasification. It offers the highest composition of hydrogen (Parthasarathy and Narayanan, 2014). Numerous studies have reported enhanced syngas yield and carbon conversion efficiency when steam was utilized as a gasifying agent (Howaniec et al., 2011; Moghadam et al., 2014; Naqvi et., 2016).

Consequently, the enhancement in pretreated material characteristics improved the gasification performance and hydrogen production (Chen et al., 2013). Thus, the co-gasification of pretreated PKS and MB coal is categorically novel in this area. The main objective of this research was to explore the influence of pretreated PKS and MB on co-gasification. The influence on co-gasification was discovered in terms of product yields, namely char, tar, gas and gases composition.

2. Methods

2.1. Materials

PKS as a biomass sample was supplied from an oil palm mill located in Penang, Malaysia. MB, which is categorized as low-rank sub-bituminous coal from Sarawak, Malaysia, was used as the coal sample. Both samples were ground and separated through a finer screen to get particle sizes in the range of 200 µm to 400 µm. The untreated PKS (PKS_{Un}) and untreated MB (MB_{Un}) were dried in at a temperature of 105°C for 24 hours and stored in an air-tight container for further analyses and pretreatment.

The pretreated or torrefied PKS (PKS_{To}) was produced via torrefaction at a temperature of 270°C with a holding time of 1 h, and the preheated MB (MB_{Pr}) was produced via preheating at a temperature of 250°C with a holding time of 1 h using fixed bed reactor. Both PKS_{To} and MB_{Pr} were prepared and selected according to our previous work (Ahmad et al., 2019). The PKS_{To} and MB_{Pr} presented an enhancement on fuel properties in terms of high calorific value, energy density, carbon content with low moisture and oxygen content at selected pretreatment temperatures (Ahmad et al., 2019). The proximate and ultimate analyses were carried out using Mettler Toledo TGA/DSC thermogravimetric analyzer and Leco CHNS-932 elemental analyzer, respectively. The calorific value was determined by using Leco AC-350 bomb calorimeter. The characteristics of all samples are shown in Table 1. The dolomite was used as the catalyst during catalytic co-gasification. The dolomite was sieved to obtain a fraction with a particle size of 45 µm. It was calcined at a temperature of 900°C for 5 h.

Table 1 Characteristics of PKS and MB

Analysis	PKS_{Un}	PKS_{To}	MB_{Un}	MB_{Pr}
Proximate analysis (wt.%)				
Moisture	10.60	1.58	21.50	4.78
Volatile matter	77.54	46.99	38.00	24.57
Fixed carbon	10.95	49.13	35.60	64.17
Ash	0.91	2.30	4.90	6.48
Ultimate analysis (wt.%)				
Carbon	47.67	52.34	54.52	59.50
Hydrogen	5.52	3.98	4.75	4.16
Nitrogen	0.39	0.90	0.97	1.00
Sulphur	0	0	0	0
Oxygen (calculated by difference)	46.42	42.78	39.76	35.34
Calorific value (MJ/kg)	18.20	20.90	20.10	23.80

2.2. Co-gasification

The co-gasification experiments were carried out using a fixed bed reactor with an internal diameter of 6 cm and a height of 30 cm at atmospheric pressure. The reactor was heated using an electric furnace. Figure 1 shows the schematic diagram of co-gasification experimental setup. Four different blending samples were used during co-gasification: (i) untreated PKS/MB (PKS_{Un}/MB_{Un}), (ii) torrefied PKS/untreated MB (PKS_{To}/MB_{Un}), (iii) torrefied PKS/preheated MB (PKS_{To}/MB_{Pr}), and (iv) catalyst-torrefied PKS/preheated MB (Cat-PKS_{To}/MB_{Pr}).

About 5 g of sample was placed inside the reactor. Initially, the reactor was purged with a nitrogen flow rate of 500 mL/min for 10 min. Then, the sample was heated to the desired gasification temperature with a heating rate of 50°C/min. A nitrogen flow was continued to generate an inert atmosphere inside the reactor. After the reactor reached the desired co-gasification temperature, the steam that was produced from the steam generator flowed into the reactor, and the nitrogen flow was stopped. The steam temperature was 120°C at a pressure of 2 bar. The steam co-gasification was held for 60 min. The co-gasification was carried out using optimization condition at a gasification temperature of 767°C, biomass blending ratio of 52% and steam flow rate of 55 mL/min based on our previous studies (Ahmad et al., 2018). These optimized conditions were used as resulting in maximum gas yield, minimum char and tar yield during the co-gasification of PKS_{To}/MB_{Pr}. In catalytic co-gasification, 10% of calcined dolomite was mixed with the PKS_{To}/MB_{Pr}.

The volatile product that left the reactor from the upper side was condensed in a tar trap. The tar trap consisted of two bottles placed in the ice bath. The non-condensable gases passed through cotton wool and silica gel to remove the remaining moisture. Then, the gas was collected in a gas bag every 15 min from the starting of steam co-gasification. When the process was completed, the furnace was switched off and the reactor was left to cool to the ambient temperature. The char product was weighted once it reached room temperature. The tar product was also measured. The gas yield was calculated by difference based on the total mass balances considering the tar and char yields. The gas analysis was carried out using Agilent 6890N and gas chromatography with a thermal conductivity detector (GC-TCD). The produced gases, namely H_2, CO, CO_2 and CH_4, were quantitatively analyzed using purified Ar as the carrier gas.

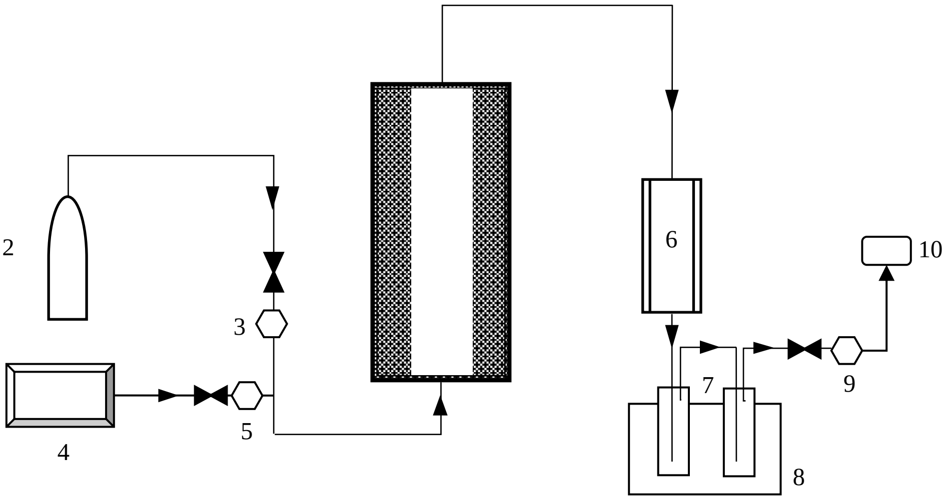


Figure 1 Schematic diagram of co-gasification experimental setup: (1) Fix-bed reactor, (2) N_2 tank, (3) N_2 flow meter, (4) Steam generator, (5) Steam flow meter, (6) Condenser, (7) Tar trap, (8) Ice bath, (9) Outlet gas flow meter, (10) Gas bag

The reaction mechanisms, such as pyrolysis reaction (Equation 1), tar cracking and reforming reaction (Equation 2), water–gas reaction (Equation 3), water-gas shift reaction (Equation 4), methanation reaction (Equation 5), steam reforming reaction (Equation 6), and Boudouard reaction (Equation 7), were involved during co-gasification.

$$\text{Biomass}_{(s)} + \text{Coal}_{(s)} \rightarrow C_{\text{biomass+coal}\,(s)} + \text{Tar}_{(l)} + \text{Gases} \quad (1)$$

$$\text{Tar}_{(l)} + H_2O_{(g)} \rightarrow \text{Gases}\ (H_2, CO, CO_2, CH_4, C_nH_m) \quad (2)$$

$$C_{(s)} + H_2O_{(g)} \rightarrow CO_{(g)} + H_{2\,(g)} \quad (3)$$

$$CO_{(g)} + H_2O_{(g)} \rightarrow CO_{2\,(g)} + H_{2\,(g)} \quad (4)$$

$$C_{(s)} + 2H_{2\,(g)} \rightarrow CH_{4\,(g)} \quad (5)$$

$$CH_{4\,(g)} + H_2O_{(g)} \rightarrow CO_{(g)} + 3H_{2\,(g)} \quad (6)$$

$$C_{(s)} + CO_{2\,(g)} \rightarrow 2CO_{(g)} \quad (7)$$

3. Results and Discussion

3.1. *Effect of Co-gasification on Product Yield*

Figure 2 presents the results obtained in terms of product yield, namely char, tar and gas. The co-gasification of PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} produced higher gas yield than co-gasification of PKS_{Un}/MB_{Un} and PKS_{To}/MB_{Un}. The gas yield increased by about 32.2% and 33.3% using PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} samples, respectively, more than using the PKS_{Un}/MB_{Un} sample. The pretreated sample exhibited a notable impact on gas production to produce higher gas yield than the untreated sample. The Cat-PKS_{To}/MB_{Pr} displays a minor increase in gas yield where only 1.1% is different from PKS_{To}/MB_{Pr} sample. Gas production using both pretreated samples was higher than Berrueco et al. (2014), which increased by only 7% more than the untreated sample on gasification of pretreated Norwegian forest residue. Moreover, the gasification of pretreated Norwegian forest residues with dolomite catalyst also showed little increase of 2% on gas yield (Berrueco et al., 2014).

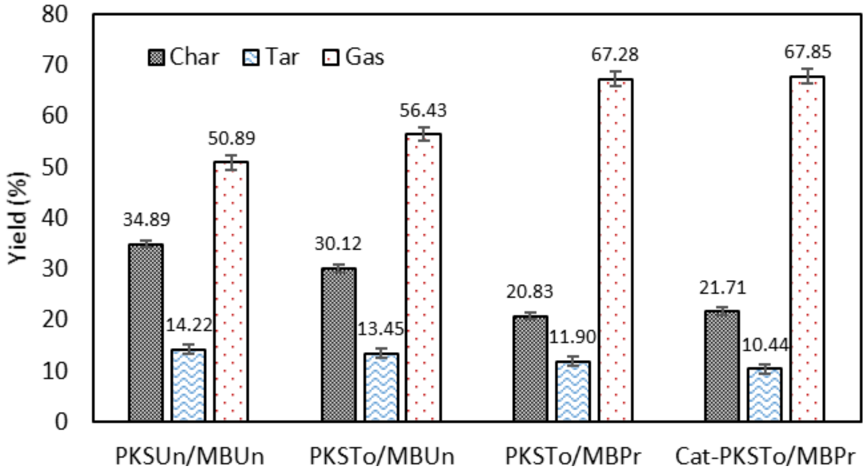


Figure 2 Effect of co-gasification of PKS_{Un}/MB_{Un}, PKS_{To}/MB_{Un}, PKS_{To}/MB_{Pr}, and Cat-PKS_{To}/MB_{Pr} on product yield

The tar yield reduced from 14.2% to 11.9% and 14.2% to 10.4% for PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} samples, respectively, which is more than for PKS_{Un}/MB_{Un}. The Cat-PKS_{To}/MB_{Pr} sample further reduced by 1.5% more than the PKS_{To}/MB_{Pr} sample for tar yield. Low tar yield is necessary for co-gasification. Thus, using both pretreated samples had noticeably reduced the tar yield in co-gasification as a significance of the partial removal of the oxygenated components of biomass and low-rank coal and volatiles through the pretreatment process. Moreover, the addition of a catalyst to both pretreated samples promotes tar reduction. These results confirm the catalytic effect of calcined dolomite that converted the tar in-situ and therefore reduced the tar yield (Berrueco et al., 2014). However, the decreasing of tar yield was not extensive when the catalyst was used together with the pretreated sample. This fact showed that in this study, using only PKS_{To}/MB_{Pr} was enough to reduce the tar.

The char yield decreased about 40.3% and 37.8% for PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr}, respectively, which is more than for the PKS_{Un}/MB_{Un} sample. The low char yield for PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} samples was associated with the increase of conversion to gas product using pretreated feedstock. Moreover, this occurrence, which was affected by the low moisture and oxygenated compound of pretreated feedstock, as presented in Table 1, made it comforting to be used as a solid fuel through co-gasification. The use of Cat-PKS_{To}/MB_{Pr} sample did not show any significant difference from the use of the PKS_{To}/MB_{Pr}

sample.

Therefore, the issue of the disadvantages characteristic of untreated biomass and low-rank coal, such as low carbon and high moisture and oxygenated content, had been overcome through the pretreatment process, which resulted in the improvement of gasification products. Additionally, this revealed that using both pretreated samples without the catalyst had extensively improved the co-gasification product yield with increasing gas yield while decreasing tar and char yield. However, the effect of the catalyst on the product yield can be seen by the optimization of co-gasification parameter, which comprises the catalyst factor.

3.2. *Effect of Co-gasification on Gases Composition*

Figure 3 shows the effect of co-gasification of PKS_{Un}/MB_{Un}, PKS_{To}/MB_{Un}, PKS_{To}/MB_{Pr}, and Cat-PKS_{To}/MB_{Pr} on gases composition at various reaction times. In Figure 3a, the H_2 composition increased with increasing reaction time for all samples. The PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} samples enhanced the H_2 production by about 63.9% and 70.7%, respectively, more than PKS_{Un}/MB_{Un} sample at 45 min reaction time. Also, the PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} increased the H_2 production by about 41% and 46.8%, respectively, more than PKS_{To}/MB_{Un} sample at 45 min reaction time.

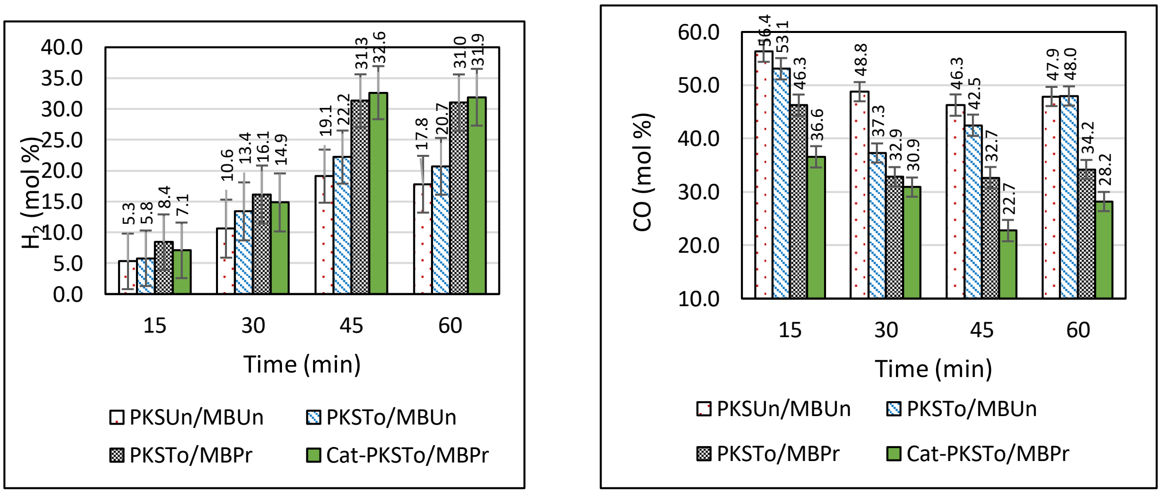
The pretreatment of biomass and low-rank coal increased carbon and calorific value, as shown in Table 1. Therefore, these had increased the energy density of the pretreated sample. As a result, more carbon had the potential to react with steam through water–gas and water-gas shift reaction during co-gasification. High production of H_2 is in accordance with the high quantity of carbon content in pretreated samples compared with the carbon content in PKS_{Un}/MB_{Un} and PKS_{To}/MB_{Un} samples. H_2 is mostly generated from the reaction of carbon with steam in the co-gasification reactor. This fact provides a great opportunity for the utilization of pretreated biomass and low-rank coal for the enrichment of H_2 production. In the case of Cat-PKS_{To}/MB_{Pr}, the addition of calcined dolomite catalyst showed a slightly higher increment than PKS_{To}/MB_{Pr} on H_2 production at longer reaction times of 45 and 60 min.

The production of CO decreased with increasing reaction time, as illustrated in Figure 3b. The PKS_{Un}/MB_{Un} blending produced a higher amount of CO than PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} blending. At 45 min reaction time, CO decreased to 46.3%, 42.5%, 32.7% and 22.7% for PKS_{Un}/MB_{Un}, PKS_{To}/MB_{Un}, PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} blendings, correspondingly. Figure 3c showed that the CO_2 content decreased as the reaction time increased. However, the Cat-PKS_{To}/MB_{Pr} sample showed a dissimilar trend for CO_2 content than other blendings. For example, the CO_2 content using Cat-PKS_{To}/MB_{Pr} blending was higher than the untreated and pretreated sample at 15 min. Then, at 30 min, the CO_2 content was lower than PKS_{Un}/MB_{Un} and PKS_{To}/MB_{Pr} blending, while at longer reaction times of 45 and 60 min, the Cat-PKS_{To}/MB_{Pr} produced the highest CO_2 content. The PKS_{To}/MB_{Pr} showed a significant reduction of CO_2 at 45 and 60 min reaction times. Figure 3d showed that the CH_4 composition increased with increasing reaction time. The PKS_{To}/MB_{Pr} and Cat-PKS_{To}/MB_{Pr} produced more CH_4 than PKS_{Un}/MB_{Un} and PKS_{To}/MB_{Un} blending. The Cat-PKS_{To}/MB_{Pr} showed the highest CH_4 production of 22.9% at 30 min reaction time, while at 45 and 60 min reaction times the PKS_{To}/MB_{Pr} produced higher CH_4 than PKS_{Un}/MB_{Un}, PKS_{To}/MB_{Un} and Cat-PKS_{To}/MB_{Pr}. CH_4 in PKS_{To}/MB_{Pr} was produced twice more than PKS_{Un}/MB_{Un} at 45 min reaction time.

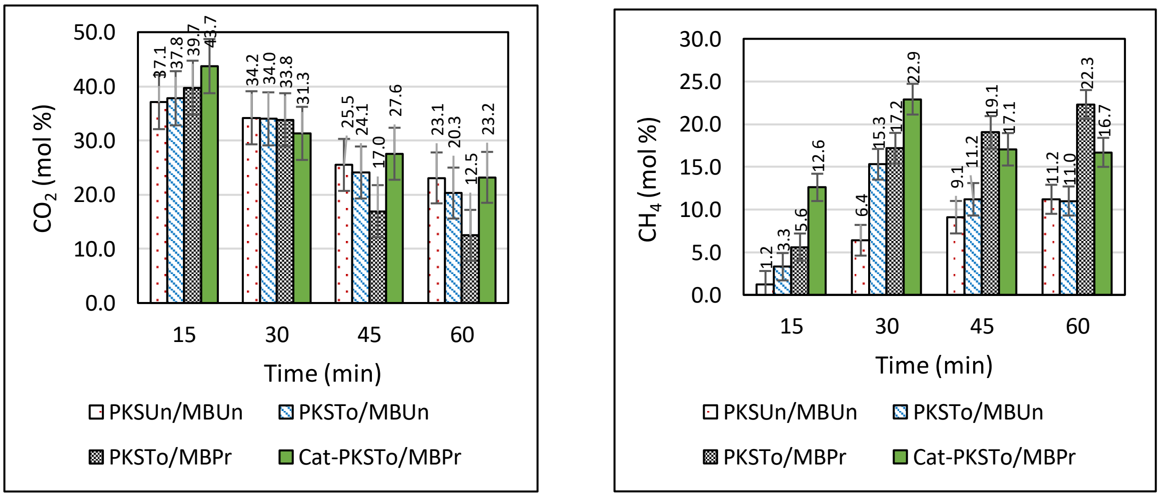
The production of gases in co-gasification involves several chemical reactions. The primary devolatisation, which is pyrolysis reaction (Equation 1) and tar cracking and reforming reaction (Equation 2), were involved in the early stage of co-gasification (Valdés et al., 2016). At 15 min reaction time, higher CO composition was produced than other gases

(H_2, CO_2 and CH_4) for PKS$_{Un}$/MB$_{Un}$, PKS$_{To}$/MB$_{Un}$, and PKS$_{To}$/MB$_{Pr}$. These indicated that the reaction, which favors the production of CO, was greater with steam gasification at 15 min. At these stages, the chemical reactions involved were preferred on water–gas reaction (Equation 3). The water–gas reaction is a reaction of carbon by steam, which can be derived from solid fuel. Cat-PKS$_{To}$/MB$_{Pr}$ produced the highest CO_2 composition, where the water-gas shift reaction (Equation 4) was dominant at 15 min. The water gas-shift results in an increase in the ratio of CO_2 to CO and of H_2 to CO.

At 30 and 45 min, H_2 and CH_4 compositions showed increment for all samples. These were directed from the water–gas reaction (Equation 3), methanation reaction (Equation 5) and steam reforming reaction (Equation 6). The highest CO composition was produced for PKS$_{Un}$/MB$_{Un}$, blending at 30 and 45 min due to the prominence of the Boudouard reaction (Equation 7) at this time. Bourdourd reaction is the reaction of the char with CO_2 during gasification. However, at longer reaction times of 45 and 60 min, the production of H_2 and CO become dominant for PKS$_{To}$/MB$_{Pr}$ due to water–gas reaction, steam reforming reaction and Boudouard reaction. For Cat-PKS$_{To}$/MB$_{Pr}$, the H_2 and CO_2 production were favored due to the steam-reforming reaction and water-gas shift reaction.



(a) H_2 (b) CO



(c) CO_2 (d) CH_4

Figure 3 Effect of co-gasification of PKS$_{Un}$/MB$_{Un}$, PKS$_{To}$/MB$_{Un}$, PKS$_{To}$/MB$_{Pr}$, and Cat-PKS$_{To}$/MB$_{Pr}$ on gases composition

Based on Figure 3, H_2 and CH_4 composition for PKS_{Un}/MB_{Un}, PKS_{To}/MB_{Un}, and PKS_{To}/MB_{Pr} increase with increasing reaction time due to water gas–reaction (Equation 3), methanation reaction (Equation 5) and steam reforming reaction (Equation 6). The composition of CO and CO_2 decreases with increasing reaction time due to water-gas shift reaction (Equation 4) and Bourdard reaction (Equation 7), which occurs simultaneously during co-gasification. Generally, for the PKS_{Un}/MB_{Un}, most atomic hydrogen is transformed into H_2O, so the concentration of H_2 is very low (Chen et al., 2013). When the PKS_{To}/MB_{Pr} is gasified, the H_2 composition rises significantly. These results clearly indicate that the pretreatment on both PKS and MB coal can facilitate H_2 production during co-gasification.

4. Conclusions

Co-gasification of PKS and MB coal was done in a fixed bed reactor. The pretreatment of a blend of both samples produced a higher gas yield with lower tar and char yield than the untreated blend of both samples. The PKS_{To}/MB_{Pr} produced a higher H_2 composition of 31.3%, which was more than PKS_{Un}/MB_{Un} sample of 19.1% at 45 min reaction time. The Cat-PKS_{To}/MB_{Pr} showed a minor increase on H_2 composition of 32.6%, which is more than PKS_{To}/MB_{Pr} of 31.3% at 45 min reaction time. The lowest CO_2 composition at 12.5% was produced by PKS_{To}/MB_{Pr} compared with PKS_{Un}/MB_{Un} at 20.3% in a reaction time of 60 min. Thus, the PKS_{To}/MB_{Pr}, which had been enriched in their properties, improved the co-gasification performance in terms of product yield and gas composition.

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