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Effect of Temperature on Catalytic Steam Co-Gasification of Rubber Seed Shells and Plastic HDPE Residues

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The catalytic co-gasification of rubber seed shell (RSS) and high density polyethylene (HDPE) mixture is investigated in a bench scale fixed bed system known as high throughput micro reactors (HTMR) system at different reactor temperature of 600 - 800 °C in order to study the effects on performance parameters such as gas and char yield, syngas yield and product gas composition. The operating temperature is selected based on the optimum conditions found from the previous study on the thermogravimetric analysis equipment coupled with mass spectrometer using these feedstock. The constant process variables used in the HTMR system are RSS particle size range of 0.100 - 0.150 mm, HDPE particle size range of 0.355 - 0.500 mm, and percentage of HDPE in the mixture of 20 weight%. The feeding rate of 2 g/h is carried out in the system.

1. Introduction

Co-gasification is one of the alternative means for generating renewable energy source, replacing the diminishing fossil fuels, overcoming the greenhouse gases emissions, and as well increasing development of waste processing technology specifically for plastic and biomass waste. A considerable amount of studies have been published in work related to the gasification behaviour of individual components of plastic and biomass waste. Meanwhile, limited studies have been focused on the binary mixtures of plastic and biomass waste in gasification process. Conversely, no studies have been conducted on catalytic co-gasification of plastic waste mixtures in a fixed bed gasifier. Fixed bed gasifier is able to provide the benefits of wide temperature distribution, easier-controlled gas quality and minimum particulate carryover compared to fluidized bed gasifier. In the present study, the behaviour of co-gasification of plastic and biomass waste mixtures with the presence of nickel catalyst is investigated. Rubber seed shell (RSS) is selected as lignocellulosic biomass for gasification feedstock since Malaysia is one of the major rubber growing countries. This results the immediate needs to study the potential of biomass rubber seed wastes utilization. High density polyethylene (HDPE) is selected as the plastic waste for gasification process. HDPE is considered to be the most commonly used plastics globally and is said to become the largest used commodity plastic material due to their versatility and wide applications. The consequence of the high demand of HDPE will definitely lead to a production of immense amount of plastic waste which lead to secondary pollution to the environment.

The catalytic co-gasification of rubber seed shell and HDPE mixture is investigated in a bench scale fixed bed system known as high throughput micro reactors (HTMR) system at different reactor temperature of 600 – 800 °C in order to study the effects on performance parameters such as gas and char yield, syngas yield and product gas composition. The constant process variables used in the HTMR system are rubber seed shells of particle size range of 0.100 - 0.150 mm, HDPE particle size range of 0.355 - 0.500 mm and percentage of HDPE in the mixture of 20 weight%. The feeding rate of 2 g/h is used.

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2. Experimental

2.1 Materials and Sample Preparation

The raw materials used in this work are RSS from Vegpro Trading, Malaysia and HDPE plastic from Shen Foong Plastic Industries Sdn Bhd, Malaysia. These materials are ground and sieved to a particle size ranging from 0 to 710 µm. Homogenized HDPE/RSS mixtures in a weight ratio of 0.2/0.8 are prepared. The ultimate and proximate analysis for RSS and HDPE are used in Tables 1 and 2 respectively. The ultimate and proximate analyses of the HDPE and RSS are conducted in LECO CHNS-932 elemental analyser and thermogravimetric analyser EXSTAR TG/DTA 6300 (Seiko Instrument Inc.) respectively.

Table 1: Ultimate analysis for RSS and HDPE in weight% (dry basis)

Sample	C	H	N	S	O
RSS	44.31	4.38	0.51	0.13	50.67
HDPE	81.45	12.06	0.34	0.79	5.36

Table 2: Proximate analysis for RSS and HDPE in weight% (dry basis)

Sample	Moisture Content	Volatile Matter	Fixed Carbon	Ash
RSS	8.59	80.98	6.62	3.81
HDPE	0.00	99.46	0.00	0.34

A commercial nickel powder (index number 028-002-01-4, Merck) is selected as the catalyst to be investigated in the catalytic gasification of HDPE/RSS mixture. The amount of nickel powder used in the present study is 10 % of the total weight of the HDPE/RSS mixture. The particle size of the nickel powder is in the range of ~10 µm and the properties of the catalyst are presented in Table 3.

Table 3: Nickel catalyst properties

XRF Analysis		Surface Properties	
Ni	96.79 wt%	Mean Pore Size	6.2 nm
Impurities	3.21 wt%	Pore Volume (BJH)	0.0016 cm^3g^{-1}
		BET Surface Area	0.78 m^2g^{-1}

2.2 Experimental Apparatus and Procedure

The HTMR system is a two-staged reactor system in which the feedstock is continuously introduced in the free fall reactor meanwhile the catalyst is placed in the fixed bed reactor. Figure 1 illustrates the schematic diagram of the HTMR system.

The pre-reactor is operated at superficial gas velocity and terminal velocity of 0.0024 m/s and 3.067 m/s respectively. The pre-reactor is continuously fed with RSS/HDPE mixtures at 2 g/h from the solid feeder at the top of the system before entering the cyclone. The cyclone is used to separate entrained solid particles. After the cyclone, the product gases are passed through a fixed bed catalyst reactor. There are four fixed bed catalyst reactors in the system. The second fixed bed catalyst reactor is used throughout the study. An approximately 200 mg of nickel catalyst is placed in the fixed bed catalyst reactor. The pre-reactor and fixed bed catalyst reactors are first purged with N_2 gas approximately at 100 mL/min to remove any entrapment of gases before both reactors are heated up to 600 °C at a heating rate of 20 °C/min. Steam is injected to the pre-reactor at temperature of 300 °C when the desired temperature of the pre-reactor is reached.

The product gases such as H_2, CO, CO_2 and CH_4 exiting the fixed bed catalyst reactor is passed through a heat exchanger before the product gases are analysed using an online gas chromatograph (Agilent 7890A) equipped with a thermal conductivity detector (TCD) which is fitted with Molsieve 5A with Hayesep Q column. The product gases are measured in 5 min interval. All experiments lasted in a duration of 90 min.

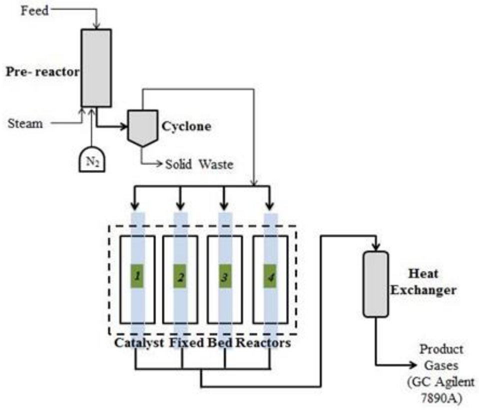

Figure 1: Experimental setup of HTMR

3. Results and Discussion

3.1 Gas and Char Yield

In the present study, the effect of temperature on the gas yield is compared with literature is presented in Figure 2. The gas yield includes the yield of the gas compositions of H_2, CO, CO_2 and CH_4. It is observed in the present study the gas yield is increased from 0.53 m^3/kg feedstock at temperature of 600 °C to 1.10 m^3/kg feedstock at temperature of 900 °C, which means an increment of 2.1 times. A similar behaviour is observed when compared with other published literature data on co-gasification of plastic and biomass (Pinto et al., 2002). Pinto et al. (2002) reported the gas yield increased from 0.63 m^3/kg feedstock at temperature of 740 °C to 1.28 m^3/kg feedstock at temperature of 880 °C in an atmospheric fluidised bed steam gasifier with cyclone and quenching system using wood pine and polyethylene mixtures as feedstock. Similarly, Brachi et al. (2014) found the gas yield increases from 0.84 m^3/kg at temperature of 651 °C to 1.30 m^3/kg feedstock at temperature of 826 °C in a pre-pilot plant steam fluidized bed with cyclone and ceramic filter using olive husk and polyethylene terephthalate (PET) in pelletized form mixtures with Ni/γ-Al_2O_3 catalyst. Hence, it can be concluded that the gas yield increases with temperature. It is further explained that the temperature enhances the main endothermic reactions such as steam methane reforming and char gasification and also volatiles during the initial pyrolysis stage (Peng et al., 2012). As a result, high temperature enhances the conversion of the feedstock to gaseous products.

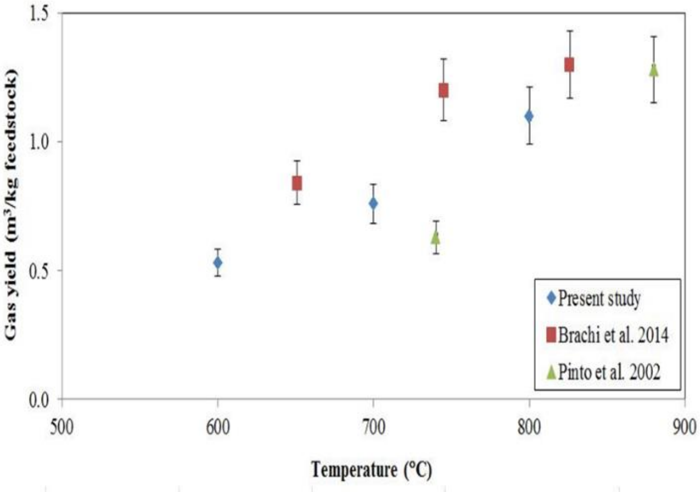

Figure 2: Effect of temperature on gas yield

The comparative effect of temperature on the char yield with literature is illustrated in Figure 3. It is observed that the char yield reduces from 12.90 g/kg feedstock at temperature of 600 °C to 6.9 g/kg feedstock at temperature of 900 °C, which means a reduction of 46.5 %. Similarly, Brachi et al. (2014) reported a decreasing trend in the char yield with temperature. However, the char yield reported by Brachi et al. (2014) is much lower than the char yield in the present study as in the mentioned temperature range. This is because Brachi et al. (2014) produce higher gas yield compared to that in the present study. The char yield reduces from 6.50 g/kg feedstock at temperature of 651°C to 2.40 g/kg feedstock at temperature of 826 °C.

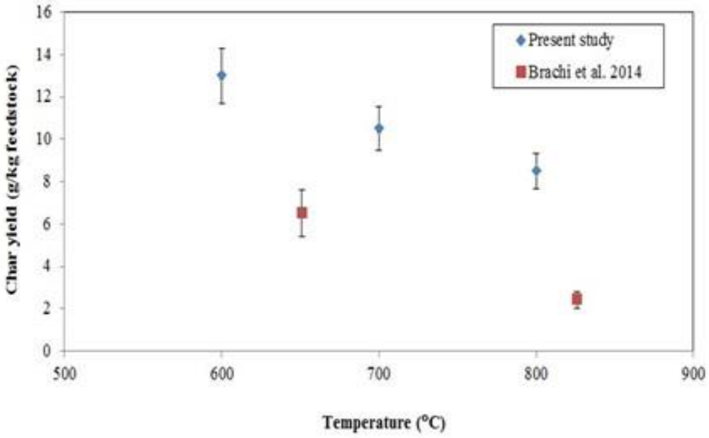

Figure 3: Effect of temperature on char yield

3.2 Syngas Yield

The effect of temperature on syngas yield is presented in Figure 4. The syngas yield includes the sum of the yield for both H_2 and CO composition. In the present study, it is observed that there is a tendency of increasing syngas yield with high temperature. A similar observation is previously reported in literature on the effect on the syngas yield with temperature (Moghadam et al., 2014). It is found that the syngas yield increases from 129 g/kg feedstock at temperature of 600 °C to 289 g/kg feedstock at temperature of 800 °C. The occurrence of this phenomenon are linked to reactions such as water-gas shift, steam methane reforming, and dry reforming reactions which are dominant during the gasification process (Midili et al., 2001). As a result, an increasing trend of syngas production yield with temperature is produced.

Results from literature on the syngas yield involving co-gasification of plastic and biomass waste mixtures are compared with the present study as displayed in Figure 4. Study reported by Moghadam et al. (2014) observed the syngas yield increased from 251.2 g/kg feedstock at temperature of 650 °C to 341.1 g/kg feedstock at temperature of 800 °C when using steam co-gasification of palm kernel shell and polyethylene mixtures. It is found that the syngas yield reported by Moghadam et al. (2014) is much higher compared to the present study. This is because a higher throughput of 2 kg/h feedstock is supplied to the pilot scale fluidized bed gasifier system in Moghadam et al. (2014) study which lead to a higher syngas yield. Syngas yield reported by Pinto et al. (2002) increased from 18.4 g/kg feedstock at temperature of 740 °C to 19.6 g/kg feedstock at temperature of 860 °C when using steam co-gasification of wood pine and polyethylene mixtures. The syngas yield reported by Pinto et al. (2002) is much lower compared to that of present study. This is likely due to the absence of catalyst in the gasification system by Pinto et al. (2002) which resulted to a lower syngas yield compared to the present study.

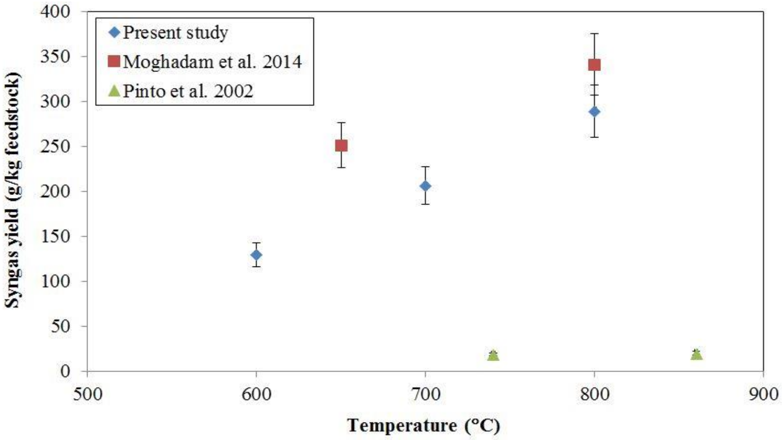

Figure 4: Effect of temperature on syngas yield

3.3 Product Gas Composition

The effect of temperature on the product gas composition such H_2, CO, CO_2 and CH_4 is shown in Figure 5. For H_2 composition, it is observed that the content increases from 38 to 51 vol% from 600 to 800 °C, which gave an increment of 34.2 %. The maximum H_2 composition of 51 vol% can be achieved at temperature of 800 °C, steam to feedstock ratio of 1.0, HDPE particle size range of 0.355-0.500 mm, RSS particle size range of 0.100-0.150 mm and percentage of plastics in the mixture of 20 wt%. This further explained that elevated temperature favours endothermic reaction such as methane reforming and tar cracking which enhanced the H_2 composition in the product gas. By varying the temperature from 600 to 800 °C, the CO content increased from 24 to 30 vol%, which referred to an increment of 0.8 times. This behaviour is attributed to the increase of the rate of heterogeneous and endothermic reactions such as water gas shift and Boudouard reactions which enhanced the formation of CO composition at higher temperature (Kim et al., 1997). Furthermore, it is seen that there is no significant effect on the concentration of CO_2 composition. This is justified that the water gas shift reaction favoured the formation of H_2 composition rather than CO_2 composition as the temperature increased. It is found that the concentration for CO_2 composition is in the range of ~10 - 13 vol% within the temperature studied. Moreover, it is also observed that the CH_4 composition decreases from 20 to 2 vol% as temperature increased from 600 to 800 °C, which correlated to a decreased of 90%. As mentioned earlier, temperature enhances the steam methane reforming reaction which lead to the consumption of CH_4.

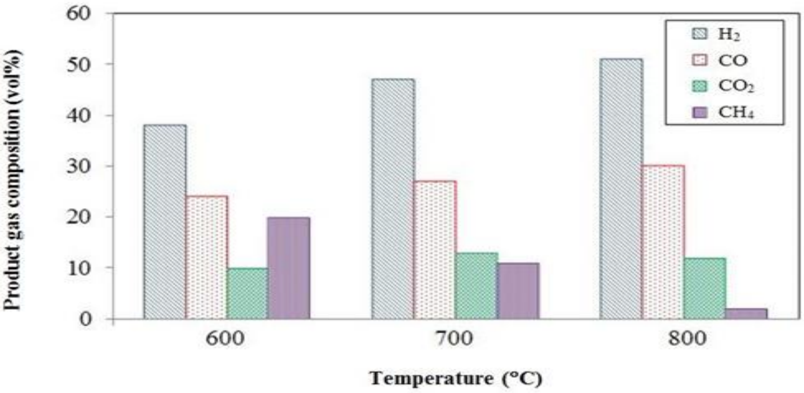

Figure 5: Effect of temperature on product gas composition

A comparative study on the effect of temperature on the product gas composition with literature is illustrated in Figure 6. In the present study, the product gas composition at temperature of 800 °C, steam to feedstock ratio of 1.0, HDPE particle size range of 0.355-0.500 mm, RSS particle size range of 0.100-0.150 mm and percentage of plastics in the mixture of 20 wt% is compared with the product gas composition obtained from the studies by Moghadam et al. (2013) and Pinto et al. (2002). It is found that the compositions for H_2, CO, CO_2 and CH_4 in the present study are 38, 24, 10 and 20 vol% respectively. Moghadam et al. (2013) carried out catalytic steam gasification in a pilot scale fluidized bed gasifier with cyclone and scrubber system at temperature of 800 °C, steam to feedstock ratio of 1.0 and polyethylene to palm kernel shell ratio of 0.3. It is reported that the gas compositions of H_2, CO, CO_2 and CH_4 obtained are 70, 7, 5 and 18 vol% respectively at mentioned operating conditions (Moghadam et al., 2013). It is observed clearly that the study reported by Moghadam et al. (2013) produced the highest H_2 concentration of 70 vol% and the lowest CO concentration of 7 vol% at the respective operating conditions when compared with present study and Pinto et al. (2002). It is also seen that the CO_2 concentration of 5 vol% produced from the study by Moghadam et al. (2013) is lower than the present study. However, the CH_4 concentration in the present study is much lower compared to Moghadam et al. (2013). Pinto et al. (2002) carried out co-gasification of polyethylene and wood pine mixtures in an atmospheric fluidized bed steam gasifier with cyclone and quenching system at temperature of 870 °C, steam to feedstock ratio of 0.8 and polyethylene to wood pine ratio of 0.1. It is reported that the gas compositions of H_2, CO, CO_2 and CH_4 obtained are 44, 35, 10 and 11 vol% respectively at respective operating conditions. It is observed that the H_2 concentration is lower compared to the present study and the study reported by Moghadam et al. (2013). This is because the study involved by Pinto et al. (2002) is solely on non-catalytic gasification process meanwhile both the present study and Moghadam et al. (2013) involved the presence of catalyst in the gasification process which enhanced the H_2 composition in the product gas. However, the CO and CH_4 concentration is much higher in Pinto et al. (2002) compared to that in the present study. This is to compensate the low H_2 composition in the product gas from Pinto et al. (2002) study.

Furthermore, it is also noticeable that the CO_2 concentration obtained by Pinto et al. (2002) and the present study is almost comparable.

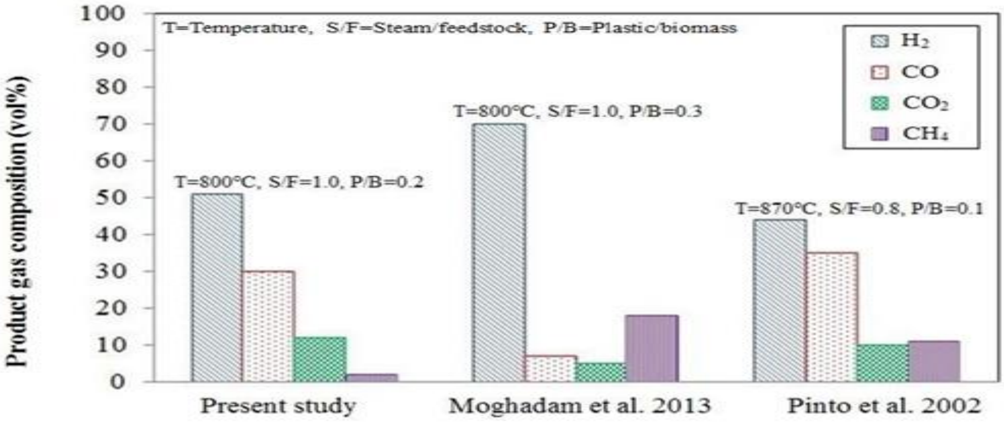

Figure 6: Comparative study of product gas composition

4. Conclusions

The parametric analysis based on different reactor temperature of 600 - 800 °C is investigated in a bench scale fixed bed reactor system i.e. high throughput micro reactors (HTMR) system. By increasing reactor temperature, it is found that the gas yield increases from 0.53 g/kg feedstock at temperature of 600 °C to 1.10 g/kg feedstock at temperature of 800 °C. And also, the syngas yield increases from 129 g/kg feedstock at temperature of 600 °C to 289 g/kg feedstock at temperature of 800 °C. However, the char yield reduces from 12.90 g/kg feedstock at temperature of 600 °C to 6.9 g/kg feedstock at temperature of 800 °C,

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