

Application of a Recycle System to Cocoa Pod Husk Gasification in a Fixed-Bed Downdraft Gasifier to Produce Low Tar Fuel Gas

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Abstract

Biomass gasification potentially generates not only producer gas but also tarry components. Practically, the gas may be used as a substitute for traditional fuel in an internal combustion engine after reducing the tar. This research examines the application of a producer gas recycle system to reduce the tar component of producer gas generated with cocoa pod husk gasification using air as a gasifying agent in a fixed-bed downdraft gasifier. The cocoa pod husk feed sizes are +1'' sieve, -1''+0.5'' sieve and -0.5'' sieve. The gasification process is operated at a temperature range of 491-940 °C and at various gasifying agent volumetric rates of 62.84, 125.68 and 188.53 NL/min or at an equivalent ratio range of 0.014–0.042. The recycle system of outlet producer gas to gasifier is set at volumetric rates of 0.139, 0.196 and 0.240 L/min. The performance of the system is evaluated by analysing the tar component using the gravimetric method of ASTM D5068-13. The gas components of CO, H₂, CO₂ and CH₄ compositions in the producer gas are also analysed with gas chromatography. This recycle system succeeded in reducing the tar content by as much as 97.19% at 0.139 L/min of the recycle volumetric rate and at a biomass feed size of -1"+0.5" sieve. The producer gas contains CO, H₂, CO₂ and CH₄ at 23.29%, 2.66%, 13.30% and 14.18%, respectively. The cold gas efficiency of the recycle efficiency is 65.24% at a gasifying agent volumetric rate of 188.53 L/min and at a biomass feed size of +1'' sieve.

Keywords: biomass gasification, cocoa pod husks, producer gas, recycle system, tar

1. Introduction

Biomass is an attractive future energy resource as its availability is sustainable and it is considered as carbon neutral. As a tropical archipelagic country, Indonesia is rich in biomass sources and every region in this country has its own specific biomass. Cocoa beans are an important commodity in Indonesia in addition to crude palm oil. Cocoa bean processing generates cocoa pod husks, which potential biomass are energy resources. Cocoa fruit consists of 70-80% of pod husks, bean shells and pulp (Vásquez et al., 2019), with one ton of dry cocoa beans generating ten tons of wet cocoa pod husks (Campos-Vega et al., 2018).

Cocoa pod husks are mainly utilised as raw material mixtures for livestock feed in Indonesia because of their protein, fibre, crude fat and mineral contents. By pyrolysis over iron oxide catalysts, the husks are synthesised to produce several chemical compounds, such as ketones, carboxylic acids, aldehydes, furans, heterocyclic aromatics, alkyl benzenes, phenols and benzenediols (Mansur *et al.*, 2014). Pectin as a chemical substance for gelling in the food industry can possibly be produced from cocoa pod husks (Campos-Vega *et al.*, 2018). Extraction using sugar acid treatment in an acidic solution is promoted as environmentally-friendly technology for pectin isolation (Priyangini *et al.*, 2018).

As biosorbents, cocoa pod husks exhibit absorbance characteristics for the removal of not only heavy metals, such as Cd(II), Pb(II), Cu(II) and Zn(II), from aqueous solution (Njoku, 2014), but also sodium diclofenac in waste water effluent (de Luna et al., 2017; Saucier et al., 2015). Limited research has been focused on husks as renewable energy sources because they are a lignocellulosic biomass (Ofori-Boateng et al., 2013; Dahunsi et al., 2019a; Dahunsi et al., 2019b) and have a relatively high calorific value of ~12.5-18.0 MJ/kg (Syamsiro et al., 2012; Titiloye et al., 2013; Forero-Nuñez et al., 2015; Adjin-Tetteh et al., 2018). Thermochemical conversion processes for husks, such as carbonisation (Syamsiro et al., 2012), fast pyrolysis to produce bio-oil, biochar and noncondensable gas (Adjin-Tetteh et al., 2018), and co-combustion of husk-coal pellets

(Forero-Nuñez *et al.*, 2015), have been investigated. Cocoa pod husks are accumulated sufficiently around the cocoa bean processing industry, which cuts handling and transportation costs. Most husks are wasted around the plantation area without any treatment.

The biomass gasification process using ambient air is a suitable and flexible method to convert cocoa pod husks into combustible gaseous products (producer gas), which mainly contain carbon monoxide, hydrogen, methane and nitrogen. Simultaneously, unwanted organic gaseous products, such as tar, are also generated. Analysis of this tar shows that aromatic and phenolic compounds dominate. The tar may interfere with the combustion process when the gas substitutes fossil fuel in an internal combustion engine. Thus, the tar should be isolated before use. Reducina tar means increasing the combustible gas content and cold aas efficiency of the producer gas simultaneously. The performance evaluation of gasification is based on the producer gas chemical compositions, including tar, the low heating value (LHV) of the gas and the cold gas efficiency (CGE).

Generally, two major methods are proposed to reduce tar contents. The primary method emphasises the prevention of tar formation in the gasification process, such as by using a fluidised bed gasifier at a reaction temperature of 900-1000 °C (Mohammed et al., 2011; Esfahani et al., 2012; Makwana et al., 2015) and at a proper equivalence ratio (ER) when using air as a gasifying agent (Ghani et al., 2009; Sarker et al., 2015). Instead of air, the use of steam as a gasifying agent with a steam/biomass ratio of 0.5-0.8 at 900 °C decreased the tar yield by 20% (Fremaux et al., 2015).

The formed tar in the producer gas can also be isolated using a secondary method, which applies an additional tar removal system after the gasifier reactor. A packed bed scrubber using a specific solvent, such as acetone and ethanol (Chang et al., 2011), linoleic acid (Malek et al., 2016) or biodiesel (Madav et al., 2019), has been applied to remove the tar content in the producer gas. These studies showed successful reductions in the tar content in the gas of 95%. An additional secondary method using high-temperature gas filtration and catalytic reforming of hydrocarbon gases and tar is also proposed to obtain tar-free fuel gases (Simell et al., 2014).

Instead of air, oxygen or steam, carbon dioxide can also be used as a gasifying agent to substantially increase the carbon and energy conversion efficiency and decrease the amount of tar in the producer gas. During the gasification process, carbon dioxide reacts with the char to form carbon monoxide by the Boudouard reaction. The carbon dioxide also promotes dry dealkylation and dry reforming endothermic reactions of hydrocarbon (including tars) in the producer gas to generate more CO and H₂ (Pohořelý et al., 2014: Antolini *et al.,* 2019; Shen *et al.,* 2019; Zhang et al., 2019). By using a recycle system of producer gas containing carbon dioxide into a downdraft gasifier with eucalyptus wood as feedstock, the tar content was successfully reduced by up to 91% (Jaojaruek et al., 2011). A thermodynamic analysis of a carbon dioxide recycle system also showed that this technique increased syngas production at high pressure and low temperature. The optimum CO_2/C was reported to be ~0.1-0.2 (Chaiwatanodom et al., 2014). When the carbon dioxide sources come from a carbon capture and storage system, an intensity of ~1.55 kg CO₂/kg wetbiomass generates carbon-negative power generation (Prabowo et al., 2015).

Furthermore, a recycle system on the updraft gasifier causes H_2 increases and CO decreases with increasing producer gas recycle rate and gasifying agent flowrate. Increasing the primary gasifying agent flowrate tends to increase the amount of CO and reduces the amount of H_2 produced (Surjosatyo, 2014). Using a recycle system on a bubbling fluidised-bed gasifier, a tar content was observed from 6–18 g/Nm³, which increases the efficiency of cold gas (Barisano *et al.*, 2015).

A briquette of lignite and sawdust mixture airgasification in an atmospheric downdraft gasifier at a temperature range of 740-915 °C was investigated by studying the effect of ER at a range of 0.240-0.386 (Upadhyay et al., 2018). The experiment showed that increasing the ER resulted in an increase of CO and a decrease in both CO_2 and H_2 . It was also observed that the cold gas efficiency of this experiment was 50.67-80.03% and the tar content was in the range of 516.3-565.23 mg/Nm³. By varying the ER from 0.20 to 0.29, wood was gasified in a downdraft gasifier using air as a gasifying agent, which generated a producer gas with a low heating value of 4.9–5.4 MJ/m^3 and CGE of 38–52% (Vonk et al., 2019).

This research investigates the gasification of cocoa pod husks in a downdraft fixed-bed gasifier with an air-recycle producer gas premixed supply system. In addition to the tar content, the gas composition of CO, H_2 , CO_2 and CH_4 is observed. The CGE of the system is also evaluated.

2. Method

2.1. Materials

Cocoa pod husks were obtained from a cocoa plantation of the Jumapolo sub-district in the Karanganyar District of the Central Java Province, Indonesia. A chemical composition and heating value analysis was executed by the Mineral and Coal Technology – Centre of Research and Development in the Ministry of Energy and Mineral Resources of the Republic of Indonesia (Table 1). As with other biomasses, the main component of the pod husks is volatile matter.

 Table 1. Typical chemical compositions and calorific value of cocoa pod husks.

Proximate Analysis (adb)	1
Moisture	12.66%
Volatile Matter	60.95%
Fixed Carbon	18.42%
Ash	7.97%
Ultimate Analysis	
Carbon	39.87%
Hydrogen	5.96%
Nitrogen	0.74%
Total Sulfur	0.13%
Oxygen	45.33%
GHV (MJ/kg)	15.46

(Analysed by Mineral and Coal Technology – Centre of Research and Development in the Ministry of Energy and Mineral Resources of the Republic of Indonesia, 2018)

2.2. Experimental Setup

All experiments were completed in a 3 kg/h downdraft gasifier with a throat diameter of 0.1 m and a total height of 1.0 m. A cyclone separator functioned as a dust collector. A test-burner for checking the gas formation before sampling was installed. Air as a gasifying agent was supplied into the gasifier with a 2 hp air blower.

The feedstocks sizes were classified into three groups: retaining on a 1.0'' sieve (+1.0''), passing on a 1.0'' sieve and retaining on a 0.5'' sieve (-1.0''+0.5'') and passing on 0.5'' sieve (-0.5''). Before feeding, the cocoa pod husks were air-dried for 2-3 d. The air flowrate varied at 62.84, 125.68 and 188.53 L/min for gasifying each feedstock size. Two different operation modes without recycling (No-R) and with recycling (R) of the producer gas at fractions of 7.0%, 9.0% and 12.0%

(v/v) were applied for each combination of feedstock size.

The tar content in the gas was collected in a series of impinger bottles, with four bottles filled with 50 mL of isopropyl alcohol solvent to absorb the tar in the gas and one empty bottle. The sampled gas flowed into the impinger set because of vacuum pump suction. This sampling method is in accordance with the guidelines for sampling and analysis of tar and particles in the producer gas generated with biomass gasification (ASTM D5068-13). After evaporating the solvent, the tar residue was weighed and the tar content in the gas was calculated using Equation (1):

$$tar = \frac{m_{tar,close} - m_{tar,open}}{m_{tar,close}} \times 100\%$$
(1)

where

tar : tar content, kg *m*_{tar,close} : mass of tar (without recycle), kg *m*_{tar,open} : mass of tar (with recycle), kg

The gas outlet from the impinger set was analysed with gas chromatography using a GC-2014 Shimadzu, TCD-14 sensor to detect CO, H_2 , CO_2 and CH_4 . Using these results and Equation (2), the LHV of the gas was calculated (Gu *et al.*, 2018):

$$LHV = \frac{10.79H_2 + 12.60CO + 35.86CH_4}{100}$$
(2)

where

LHV : heat value of syngas, kJ/Nm³ H₂ : mole fraction of H₂ in the gas CO : mole fraction of CO in the gas

 CH_4 : mole fraction of CH_4 in the gas Air and gas sample flowrates were measured

Air and gas sample flowrates were measured with rotameters, while the recycled gas was determined with an orifice plate. The throat temperature was monitored with a 1200 K thermocouple. Three bimetal thermometers were installed to monitor the temperature of the recycled gas, cyclone outlet gas and gas samples. All the equipment was assembled as shown in Figure 1 at the Pilot Plant and Energy Conversion Laboratory of the Faculty of Engineering, Universitas Sebelas Maret in Surakarta.

After completing the process, the solid residues were weighed for the calculation of the CGE using Equation (3). CGE is defined as the ratio between the flow of energy in gas and energy contained in biomass feedstocks. This CGE value represents the performance of the gasifier configuration used for gasifying the cocoa pod husks.



Figure 1. Experimental setup.

$$CGE = \frac{V_{gas} \times LHV_{gas}}{m_b \times LHV_b} \times 100\%$$
 (3)

where

CGE: cold gas efficiency V_{gas} : syngas flow rate, Nm³/hLHV: heating value of syngas, kJ/Nm³ m_b : mass feed biomass, kg/hLHV: heating value of biomass, kJ/kg

3. Results and Discussion

3.1. Producer Gas Composition

Based on the results (Figure 2), the concentration of CO gas shows an increasing tendency (9.55-17.49%) with the recycle system and a fluctuating condition between 4.71% and 23.29% without the recycle system. The feedstock size to obtain the highest CO concentration in the recycle system was for retaining on the 1" sieve, with an air flowrate of 188.53 L/min (ER = 0.042) and an average gasification temperature of 883 °C. In contrast, without the recycle system, the feedstock size and air flowrate to obtain the highest CO concentration were retaining on a 1" sieve and a 125.68 L/min air flow rate (ER = 0.028), respectively, and an average gasification temperature 761 °C.

Although more CO content was produced in the producer gas without the recycle system, the syngas product was lower than with the recycle system. The recycle system is capable of generating more producer gas with a relatively high CO content. The thermodynamic study of biomass gasification using CO_2 recycling proved to increase the gas production. It is obvious that the water gas shift reaction promotes CO production with the increase of CO_2 concentration in the gasifying agent (Chaiwatanodom *et al.*, 2014). Again, the Boudouard reaction also proceeded in the gasifier and it was enhanced by the increase of CO_2 injection from recycling (Prabowo *et al.*, 2015).

With recycling, the hydrogen composition tends to rise from 9.21% to 13.30% with an increasing in the air flowrate from 62.84 to 188.53 L/min (Figure 3). The highest H₂ concentration of 13.30% was obtained at an air flowrate of 188.53 L/min (ER = 0.042) and retaining on the 1" sieve feedstock size with an average temperature of 883 °C. Similar to CO_2 , a fluctuation of hydrogen composition was identified when no recycle system applied. The highest H₂ concentration is 11.67% when the air flow rate was 125.68 L/min (ER = 0.028) and the size of the feedstock passing 0.5 in sieve with an average gasification temperature of 761 °C.

The results of the present study are in accordance with the research conducted with an updraft gasifier at 1148–1273 K and biomass size of 3 cm x 3 cm (Surjosatyo et al., 2014). With increasing recycle flowrate from 0.0183 to 0.035 L/min, the obtained H_2 gas concentration increased from 11% to 17%. This is also directly proportional to the using wood pellet biomass, research bioplastic pellets and olive husk pellets as feedstocks of a fluidised bed gasifier and steam as the gasifying agent (Ruoppolo et al., 2012). The operating conditions were an ER of 0.09-0.30 at temperature of 780 °C.



Figure 2. Effect of air flowrate on CO content







Figure 4. Effect of air flowrate on CH4 content



Figure 5. Effect of air flowrate on CO₂ content

Hydrogen gas concentrations showed a tendency to increase and fluctuate as the ER value and the ratio between steams to fuel increase.

As a combustible gas component of producer gas from biomass gasification using air as the gasifying agent, methane exhibits the lowest concentration (~3%) compared with carbon monoxide and hydrogen. The maximum methane concentration of 2.66% was obtained without using the recycle system at an air flowrate of 125.68 L/min (ER = 0.028), retaining on a 0.5" sieve of feedstock size and an average gasification temperature of 771 °C. It was observed that the maximum concentration was lower (1.83%) when using the recycle system with a higher equivalent ratio at 0.042 (Figure 4).

The results agree with the research conducted in an updraft gasifier at 1148–1273 K (Surjosatyo *et al.*, 2014) and in a fluidised bed gasifier at 780 °C (Ruoppolo *et al.*, 2012). Both identified that a higher recycle flow rate increases methane in the producer gas significantly.

Figure 5 shows that the recycle system generated more CO₂ at the same air flowrate and the same feedstock size. The fraction tends to decrease with increasing air flowrate for all feedstock sizes. With the recycle system, the highest CO₂ produced was 14.83% while 13.51% was obtained without the recycle system at the same air flowrate of 62.84 L/min and ER of 0.014. At the same ER, the recycle system was able to increase gasification temperature compared to without the recycle system from 742 to 836 °C.

This condition agrees with previous research (Li *et al.*, 2004), with the CO₂ gas decreasing from 14.5% to 11.7% with an increasing of air-fuel ratio from 0.20 to 0.45 at a gasification temperature of 700–850 °C. Experiments without recycling showed a fluctuating CO₂ content in the gas. This is in accordance with research using a fixed-bed gasifier reactor at a temperature of 1000 °C and an ER of 0.02–0.87 without recycling (Yin *et al.*, 2012). With increasing the feedstock size from 1 to 8 cm, the CO₂ in the producer gas fluctuated significantly.

3.2. Tar Reduction

It is presented in Figure 6 that using producer gas recycling, which contains carbon dioxide, together with air as a gasifying agent was likely to reduce the tar content in the gas. The tar reduction increased significantly at a medium volume fraction of recycle gas (9.0%) in the gasifying agent mixture but decreased when using more fraction. Using a medium feedstock size of passed 1.0" and retained on 0.5" sieve size resulted in the highest reduction at all recycle volume fractions. However, there is only a marginal increase of tar reduction from 92.0% to 97.2% when using this size at a higher volume fraction.

In general, the observed gasifier temperatures were higher at a low volume fraction of recycle gas rather than at a high volume fraction. The possible explanation is that more CO_2 content in the gasifying agent mixture promotes endothermic boudouard and dry dealkylation reactions, with potential to decrease the temperature.



Figure 6. Effect of recycle gas in gasifying agent on tar reduction at different feedstock sizes



Figure 7. Effect of air flowrate on CGE

This is consistent with previous research using wood as fuel for gasification in a fluidised bed gasifier (Pohořelý et al., 2014). It is also reported that the tar content in producer gas reduced to less than 45 mg/Nm³ when using downdraft gasifier with innovative two-stage air and a premixed air/gas supply (Jaojaruek et al., 2011). This gas is possible to be injected directly into an internal combustion engine. A previous research using the same fixed-bed downdraft gasifier and palm kernel shell as feedstocks reviewed the same behaviour (Pranolo et al., 2018). The recycle gas to the gasifier reduced the tar content in the producer gas up to 62% at temperatures of 750-780 °C.

3.3. Cold Gas Efficiency

The CGE describes the ratio of calculated heating value of the producer gas using combustible contents of the gas and the heating value of biomass feedstocks, which indicates the performance of the gasification

configuration. This study shows that the recycle system of producer gas into the gasifier improves the CGE significantly by ~12%-34% at a high flowrate of gasifying agent (Figure 7). Even more when using a smaller particle size, the recycle system improves the CGE significantly. At a medium air flowrate of 125.68 L/min, a gasifying smaller feedstock size (retaining on 1.0" sieve) resulted higher CGE compared with higher air flowrate, but this generates less producer gas. This result agrees with previous work using two-stage air-gas supply which improved gas efficiency and the capacity around 15% and 40%, respectively (Jaojaruek et al., 2011).

A previous research also confirmed that increasing the air flowrate resulted increasing gasification efficiency in an updraft gasifier. Using an updraft gasifier without recycling for coconut shell gasification, increasing the air flowrate from 70.2 to 122.4 L/min caused the gasification efficiency raised from 40% to 55% (Vidian, 2008). The efficiency would continue to rise until a certain maximum point, then would decrease as the combustion air flowrate increases. Further increasing the air flowrate causes low combustible gas content in the producer gas because the combustion that occurs is more perfect.

4. Conclusion

Application of a recycle system to cocoa pod husk gasification in a fixed-bed downdraft gasifier reduces tar content in the gas significantly. This also improves the CGE more when a smaller particle size of cocoa pod husk is applied. Carbon monoxide and hydrogen content of the gas are in the acceptable range as fuel gas. Thus, the gas may substitute the diesel fuel partially for generating electricity.

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