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Effect of water addition in a microwave assisted thermal cracking of biomass tar gasification

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Abstract. Producer gas from biomass gasification is plagued by the presence of tar which causes pipe blockages. Thermal and catalytic treatments in a microwave reactor have been shown to be effective methods for removing tar from producer gas. A question arises as to the possibility of enhancing the removal mechanism by adding water into the reactor. Thermal treatment with a various amount of water was added at temperatures in the range of 800–1200°C. The tar removal efficiency obtained 96.32% at the optimum temperature of 1200°C at the water to tar ratio (W/T) of 0.3. This study shows that the removal of tar by microwave irradiation with water addition is a significant and effective method in tar cracking.

1. Introduction

Biomass gasification is an efficient and environmentally friendly method to produce gaseous fuel (producer gas) for heat and power generation through internal combustion engine, gas turbine, and fuel cell as well as for chemical synthesis applications such as methanol, methane and Fischer–Tropsch liquids. However, producer gas is always accompanied by undesirable products such as tar and particulates. Until now, tar presents the main obstacle in biomass gasification for not only causing serious operational problems in a downstream pipeline and end-user application but also affecting the energy efficiency of the overall process. Therefore, removal of tar from producer gas is indispensable.

Since three decades ago, various producer gas cleaning methods have been developed and reported in some reports with the aim to produce high-quality producer gas for end-user application. In general, these methods are classified into two categories, namely: primary methods that consist of gasifier design and optimization of operating conditions and secondary methods that consist of mechanical and thermocatalytic treatments [1,2]. From the technical point of view, a combination of the two methods guarantees a more satisfactory quality of producer gas [3]. Moreover, appropriate implementation of thermocatalytic treatment is more favorable due to converting tar into useful gases such as hydrogen, carbon monoxide or hydrocarbon gases which can improve the producer gas energy content.

Until now, it is undeniable that thermal treatment process improves the composition of the producer gas, however, little attention has been paid to its development since it requires high additional energy to achieve the desired operating temperatures. Reports show that most of the heavy tar can be cracked at a temperature of 900 °C [4]. Even so, to achieve sufficiently high tar conversion



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efficiency, temperatures of more than 1100 °C are needed [5]. In another approach, catalytic treatment processes have gained more attention. In particular, there have been ongoing efforts for developing more economical catalysts for tar conversion. For this reason, the utilization of natural catalysts such as dolomite and zeolite or their impregnation on metal catalysts would be a wise choice.

Based on the preceding concerns, implementation of radio frequency (RF) energy for thermocatalytic treatment of tar would be a more realistic option. In this method which commonly utilizes microwave, the transfer of energy into the material occurs instantaneously through molecular interaction with the electromagnetic field [6]. The unique feature of volumetric heating of this technique results the more rapid heating process of the reactor in the presence of susceptor material [1] resulting in significant energy saving, reduce process time, increase process yield and environmental compatibility [7]. For this reason, a simple and rapid test technique has been developed by using RF energy which is not only effective but has also low energy consumption. The performances of this technique have been demonstrated and reported in our previous paper for thermocatalytic treatment of tar using toluene and naphthalene as tar model compounds [1].

This work was conducted to examine the capability of RF thermocatalytic treatment reactor in improving the quality and production of producer gas. The producer gas containing tar and particulates was continuously produced from rubber woodblocks gasification in a suction fixed bed downdraft gasifier. The effects of temperatures and catalysts (calcined dolomite and Y-zeolite) on the yield of products including tar, particles, and gas composition as well as high heating value (HHV) of producer gas were studied.

2. Materials and Methods

2.1. Materials

In this study, pellets wood as biomass material for the production of producer gas in a suction throatless downdraft fixed bed gasifier. The feed material was prepared into small pieces with a size 5 mm and 3 cm length. Proximate analysis showed that the feed material was comprised of 11.4 wt.% of fixed carbon, 78.3 wt.% of volatile matter, 0.2 wt.% of ash and 10.29 wt.% of moisture. Whilst elemental analysis revealed that it was composed of 44.80 wt.% of carbon, 12.19 wt.% of hydrogen, 0.45 wt.% of nitrogen, 0.88 wt.% of sulfur and 41.68 wt.% of oxygen. The HHV of the feed material determined using an automatic bomb calorimeter was 20.6 MJ kg⁻¹.

2.2. Experimental apparatus

The schematic of overall experimental setup is given in supplementary material (Fig. 1). It consists of three main units: gasifier, producer gas treatment, and gasification product collection systems. The system includes a suction throatless downdraft fixed bed gasifier, cyclone separator, condenser, blower, and a flare port. The gasifier with an inner diameter of 0.15 m and height of 1.05 m has a thermal power output of 10 kW corresponding to 6 kg·h⁻¹ biomass feeding rate. An orifice meter with a differential pressure transducer was used to measure the pressure drop for flow rate quantification of the producer gas.

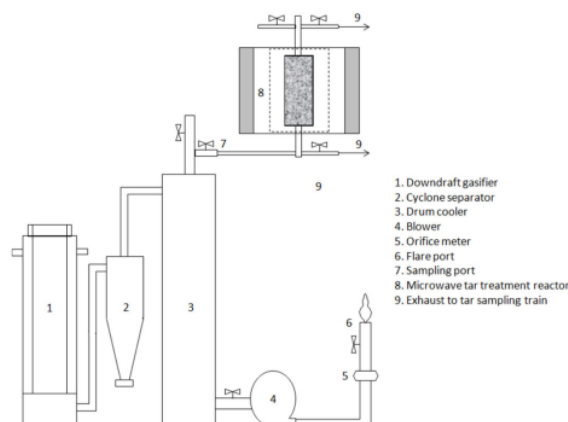


Figure 1. A schematic of the experimental apparatus for tar conversion

Producer gas treatment system includes a modified RF oven and a reactor containing silicon carbide (SiC) as susceptor material. The modified RF oven (Panasonic, NN-SM330 M) has a frequency of 2.45 GHz corresponding to wave-length of 12.23cm. The maximum power consumption and maximum output power of the RF oven were 1125 and 700 W, respectively. The alumina reactor (25.4mm i.d. and 160 mm length) was installed vertically in the RF chamber and designed as a fixed bed reactor. To absorb and convert RF energy into heat, SiC with a particle size of 2.085 mm and bulk density of 1.48 g cm^{-3} was employed. This system is also called RF tar treatment system and has been fully described in a previous study [1]. For maintaining the temperature inside the reactor, a temperature controller was also attached to the system.

Gasification product collection system consists of a tar sampling train, flow meter, vacuum pump and a gas sampling bag. The tar sampling train has been modified along with the guideline for sampling and analysis of tar and particles in producer gas [8]. The producer gas passes through a series of six impinger bottles. The first two bottles were placed in an atmospheric environment and the second four bottles were immersed in a mixture of ice and salt bath with a temperature of about $-22 \text{ }^{\circ}\text{C}$. In addition, the first five bottles were filled with 50mL isopropanol whilst the last was empty. The consideration of the modified tar sampling train is based on the behavior of tar produced by a downdraft gasifier that is mainly composed of class 3 and 4 tar [9]. The vacuum pump was used to extract the producer gas into the RF tar treatment system. A flow meter measures the flow rate of dry clean producer gas through the RF reactor and gasification product collection system.

2.3. Experimental procedure

2.3.1. Thermal treatment

For each experiment, a portion of the raw producer gas generated from pellets wood gasification was taken after the condenser (see Fig. 1) and then extracted into the RF tar treatment fixed bed reactor at a particular flow rate to give the desired residence time. The rest of the raw producer gas was burned on the flare port. In thermal treatment, the temperature was varied from 900 to $1200 \text{ }^{\circ}\text{C}$ whereas the residence time was in the range of 0.12 – 0.13 s. In catalytic treatment, the catalyst with SiC bed is sandwiched between SiC beds, forming a total bed height of 120 mm within the reactor.

2.3.2. Sampling and analysis of the product

Before and after leaving the RF tar treatment reactor, the gas stream was passed into a tar sampling train containing organic solvent of isopropanol to condense and absorb the tar as well as particulates.

Once the samples have been collected, they were mixed together and filtered to separate the particles through the preweighed qualitative filter paper (Whatman, 90 mm diameter) into a flask. The filter paper containing particles was dried in an oven and then weighed to obtain final weight. The difference of initial and final weight of the filter paper was considered as a yield of particles. The filtered solvent was evaporated by a standard rotary evaporator equipped with solvent library software. Gravimetric tar yield was obtained by the weight of the dry residue normalized by the collected gas volume. Tar samples were analyzed using gas chromatography - mass spectrometry (GC-MS) analyzer combined with NIST MS 2.0 software. The dry clean gas product was collected using a gas sampling bag and then analyzed in a gas chromatography-thermal conductivity detector (GC-TCD) to quantify producer gas composition using He as a carrier gas. Three samples were taken to obtain the average.

2.3.3. Calculation of equivalence ratio and gas heating value

In this work, the biomass gasification was run under a fixed condition for each experimental study. In order to reduce the number of parameters affecting the performance of the biomass gasifier, an equivalence ratio (ER) of 0.26 was maintained constant. ER reflects the combined effect of airflow rate, the rate of wood supply and duration of the run that can be calculated based on the following equation [1]:

$$ER = \frac{\text{Air flow rate}}{\text{Biomass consumption rate}} / \frac{\text{air flow rate}}{\text{biomass consumption rate}} | \text{Stoichiometric} \quad (1)$$

The stoichiometric ratio of air flow rate to biomass consumption rate is 5.22 m³ air/kg of wood [1].

Evaluations of the gasification performance such as gas yield, carbon conversion efficiency, and cold gas efficiency were excluded because only a fraction of the producer gas can be processed in the RF reactor. Thus, in this study, the producer gas quality was characterized by the tar and particles contents, producer gas compositions and high heating values (HHV) of the producer gas. HHV (MJ Nm⁻³) of the producer gas is dependent on the percentage volume fraction (X) of H₂, CO, and CH₄ and can be calculated by the following equation:

$$HHV = 12.766X_{H_2} + 12.6441X_{CO} + 39.847X_{CH_4} \quad (2)$$

3. Results and Discussion

Since the raw producer gas is taken after the condenser and directly introduced into the RF reactor without gas pre-filter, it contains not only producer gas (H₂, O₂, N₂, CO, CH₄, and CO₂) and tar but also particulates and water. The result showed that the raw producer gas consisted of 94.25 wt.% of gases, 0.19 wt.% of tar, 0.04 wt.% of particles and 5.51 wt.% of water. Meanwhile, the producer gas was mainly composed of 12.96 vol.% of H₂, 16.67 vol.% of CO, 1.77 vol.% of CH₄, 12.89 vol.% of CO₂, 3.16 vol.% of O₂ and 52.55 vol.% of N₂. Consequently, several reactions take place simultaneously during thermocatalytic treatment of producer gas tar in the RF reactor.

Thermal treatment of tar

Gas compositions and HHV of producer gas from thermal treatment process at various temperatures are shown in Figure 2. H₂ content seems to decrease from the initial condition after thermal treatment at 900°C. Hydrocracking of tar and hydrogasification of solid particles take a more prominent role at this condition that consumes more H₂, resulting in increasing CH₄ production.

The increase of CO formation is mainly formed by partial oxidation of solid particles. Fig. 2 displays the yields of tar and particle as a function of temperature. More than 85% of the particles were consumed and converted into gases whilst tar conversion reached only about 50% at 900°C showing less contribution of tar cracking and reforming reactions for H₂ production. Compared to other similar studies, this system resulted in lower tar conversion efficiency due to the presence of light aromatic and light poly-aromatic hydrocarbon which is relatively stable.

The gas composition shows different behavior at higher temperatures where the formation of combustible gases become more intense. Thermodynamically, when the reaction temperature increases, CO, and H₂ formation increases whilst CO₂ and CH₄ formation decreases. The presences of O₂, H₂O, and particles support the production of combustible gases by means of water gas reaction, Boudouard reaction, and partial oxidation.

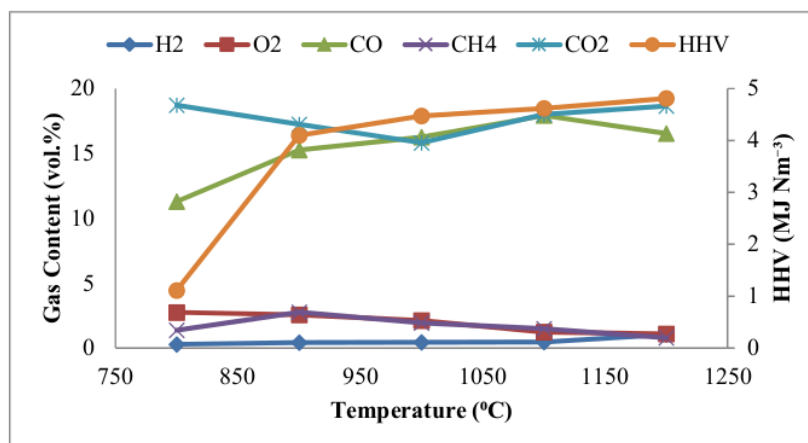


Figure 2. Tar and particle concentration with thermal treatment of producer gas

According to Le Chatelier's principle, the high reaction temperatures favor the first two endothermic reactions. Moreover, the conditions also occur due to the high activity of tar cracking and reforming reactions. In the meantime, the CH₄ content shows a slight improvement from 900 to 1000 °C and then decreases as the reaction temperature increases. The reduction of CH₄ content at higher temperature is mainly caused by steam and dry methane reforming reactions. These reactions are also endothermic, meaning the forward reaction is preferred at higher temperatures. As a result, the gas heating value is enhanced by about 18% at the highest reaction temperature of 1200 °C. More than 90% and 98% tar and particle conversion efficiencies were obtained, respectively. These results correspond to the tar and particle concentration of 147 and 4 mg Nm⁻³, respectively. This achievement is in line with other studies where complete conversion of tar and particle in the producer gas requires extremely high temperatures above 1200 °C. Tar compounds identified by GC-MS from raw producer gas and after thermal treatment processes of producer gas tar are shown in Table 1. Tar species contained in raw producer gas generated by downdraft gasification of rubber woodblocks include heterocyclic compounds (e.g., phenol and benzofuran), aromatic compounds (e.g., benzene, toluene, xylene, ethylbenzene, styrene, indene and methylidene), light poly-aromatic hydrocarbons (PAHs) (e.g., naphthalene, methylnaphthalene, fluorene and anthracene) and heavy PAHs (e.g., pyrene, fluoranthene, chrysene and acepyrene). It can be noted that these compounds are commonly identified in the case of biomass downdraft gasification [8,9].

The identified tar compounds changed considerably when the producer gas tar was allowed to pass within the high temperature of RF reactor. Some of the tar species especially aromatic compounds (e.g. xylene, ethylbenzene, and styrene), light PAHs (e.g. methylnaphthalene, fluorene and phenanthrene) and heavy PAHs (e.g. benzo[b]fluorine, chrysene, and acepyrene) were not detected during thermal treatment at 900 °C. Increasing the reaction temperature up to 1200 °C, it can be seen that only benzene, toluene, indene, naphthalene, biphenyl, and anthracene can be found in tar species. In addition, more than 70% of tar species was occupied by benzene and naphthalene. This result confirms the high stability of these compounds during thermal treatment process [1].

Table 1. Relative area percentage of major tar compounds resulting from thermal treatment of producer gas tar

Compound name	Molecular weight	Boiling point (°C)	% of compound in producer gas tar				
			Untreated	Thermal reaction temperature (°C)			
				900	1000	1100	1200
Benzene	78	80	11.01	23.97	25.06	31.64	35.52
Toluene	92	110.6	13.10	11.62	15.20	14.18	4.40
Styrene	104	145	0.40	-	-	-	-
Ethylbenzene	106	145	0.65	-	-	-	-
p-Xylene	106	138.3	1.26	-	-	-	-
o-Xylene	106	144.5	0.72	-	-	-	-
Benzene, 1,2,3-trimethyl	120	98	0.27	8.81	6.68	-	-
Benzene, (1-methylethyl)-	120	172.8	2.65	-	-	-	-
Benzene, 1-ethenyl-2-methyl-	118	172.8	1.47	-	-	-	-
Benzene, 1-ethenyl-3-methyl-	118	172.8	3.31	9.53	6.03	-	-
Benzofuran	118	174	0.26	-	-	-	-
Phenol	94	181.8	4.82	2.02	-	-	-
Indene	116	182	6.31	3.76	7.16	7.77	10.23
Methylindene	130	199	3.88	7.16	-	-	-
Naphthalene	128	217.9	4.46	13.80	23.98	35.64	34.80
2-Methylnaphthalene	142	241.1	2.19	4.57	4.33	-	-
1-Methylnaphthalene	142	244.7	2.47	3.43	4.41	-	-
4-Butyl-1,1'-biphenyl	210	318	1.46	3.74	4.03	6.23	8.48
Anthracene	178	339.9	1.32	3.14	3.10	4.54	6.57
Diphenylethyne	178	256.1	2.13	-	-	-	-
9H-Fluorene-9-methylene-	178	295	0.39	-	-	-	-
Phenanthrene	178	340	0.49	-	-	-	-
Methylenephenanthrene	190	353	2.54	-	-	-	-
Anthracene, 9-methyl-4-	192	196	1.00	-	-	-	-
Methylenephenanthrene	192	197	0.66	-	-	-	-
Fluoranthene	202	384	2.68	2.01	-	-	-
Pyrene	202	404	11.07	2.45	-	-	-
Pyrene, 4,5-dihydro-	204	404	2.92	-	-	-	-
11H-Benzo[b]fluorene	216	405	3.81	-	-	-	-
Pyrene, 1-methyl-	216	405	3.50	-	-	-	-
Chrysene	228	448	1.15	-	-	-	-
Accepyrene	226	448	5.31	-	-	-	-

4. Conclusions

The performance of RF tar thermocatalytic treatment system was investigated. Temperature plays a crucial role for tar and particle conversions under the investigated conditions. The thermal treatment

produces a higher heating value of 5.76 MJ Nm⁻³ at 1200°C. Both dolomite and Y-zeolite offer better tar and particles conversion efficiencies of around 97%. Even at a higher temperature of thermocatalytic cracking benzene, naphthalene and toluene were still found. The presence of reforming products, water, and particulates in the producer gas has a major impact on upgrading producer gas quality.

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