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Original Article

Gasification of municipal solid waste in a downdraft gasifier: Analysis of tar formation

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Abstract

In this study, municipal solid waste (MSW) from a dumpsite was converted into refuse derived fuel (RDF) and used as feedstock for an air-blown gasification process. The gasification process was conducted in a 10 kg.hr^{-1} downdraft gasifier at different air flow rates of 300, 350, 400, 450 and 550 NL.min^{-1} at atmospheric pressure in order to investigate the quantity and quality of tar formed. It was shown that the increase in the air flow rate from 300 NL.min^{-1} to 550 NL.min^{-1} led to an increase in the oxidation temperature from 719°C to 870°C and an increase in the reduction temperature from 585°C to 750°C, respectively. Tar was reduced from 15 g.Nm^{-3} to 4.7 g.Nm^{-3} respectively. Heavy tar compounds ($\geq C_{17}$) e.g. pyrene and phenathrene, decreased with the increase in the light tar compounds ($\leq C_{10}$) e.g. naphthalene, indene and toluene; indicating the presence tar reduction through a tar cracking process.

Keywords: tar formation, tar reduction, air blown gasification, downdraft gasifier

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1. Introduction

The increase in energy demand and the fossil fuels dependency as well as the depletion of fossil fuels show that it is necessary to find the alternative and sustainable sources of energy. Furthermore, use of alternative energy will help to minimize environmental problems associated with the use of fossil fuels (Gonzalez *et al*., 2008). Alternative energy sources include biomass, solar, wind, geothermal, and tidal wave. Each form of alternative energy is effectively used in different parts of the world based on political willingness, available potential, technology available and accessible markets and others (IEA, 2001; IEA, 2003). In Thailand, biomass is one of the available and promoted alternative energy sources. Biomass as an energy carrier includes virgin wood, energy crops, agricultural wastes, food wastes, industrial wastes and co-products (BEC, 2011). Municipal solid waste (MSW) is one form of biomass. These wastes include kitchen waste, garden waste, plastic waste, paper waste and many others (EPA, 2012). Fresh MSW has high moisture and low energy content, contrary to old MSW that was disposed on landfill. Landfill reclamation is needed to reclaim land for reuse and recovery of fuel fraction that could be used for energy production. Treatment or upgrading of fuel fraction from reclaimed landfill is necessary to obtain high quality fuel. The treatment involves reclaiming of combustible wastes from the rest of waste, which are then processed into compacted fuels (refuse derived fuel, RDF) suitable for thermal conversion.

Regarding of thermal conversion pathways of RDF to green and clean energy, gasification is considered as promising option (Knoef, 2005; Zaman, 2009; Lei *et al*., 2011). It is the partial oxidation of carbon containing fuels by the

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use of gasification agent into mixture of combustible gases at temperatures above 500°C. The gasification agent can be air, oxygen, steam or carbon dioxide. The gas produced from gasification process is called producer gas and it contains mainly of carbon monoxide, hydrogen, methane, and light hydrocarbons (Knoef, 2005; Lei *et al.*, 2011).

Studies reported that gasification of MSW using fixed bed gasifiers has a potential for small scale power production (<1MW) and it is feasible to produce 15-150 MW using fluidized bed gasifiers (Knoef, 2005). However, the main drawback of gasification is the presence of tar, char, and particulates in producer gas (Klein, 2002; Murphya and McKeoghb, 2004; Knoef, 2005; Jenkins, 2007; Anis and Zainal, 2011; Kaewluan and Pipatmanomai, 2011; Ahmed *et al.*, 2013).

Tars are hydrocarbons with molecular weight more than benzene, which condense at low temperatures outside the gasifier. Tars compound include phenol, toluene, styrene, xylene, indene, naphthalene, phenanthrene and pyrene (Evans and Milne, 1997; Phuphuakrat *et al.*, 2010; Siedlecki and de Jong, 2011). For example, during power generation, tars in producer gas may condense and deposit leading to blocking of pipes, filters, and valves; decreasing machine performance while increasing maintenance cost (Milne *et al.*, 1998; Knoef, 2005; Anis and Zainal, 2011). Power generation equipment, e.g. gas turbine and gas engine, have limitations for tar concentration, as shown in Table 1.

However, different gasifier types produce different amount of tar. Downdraft gasifiers produce lower amounts of tar compared to other gasifiers, as shown in Table 2. Despite the low amount of tar from downdraft gasifier, the tar quantities are above limitations, therefore a method of tar removal is necessary.

Tar removal methods are grouped into primary and secondary methods depending on where tar removal takes place. In primary methods, tars are removed within a gasifier by optimizing gasification parameters, e.g. air supply, temperatures and use of catalysts. Secondary methods involved tar removal outside the gasifier via physical treatment e.g. scrubber and filters (Devi *et al.*, 2003; Abu *et al.*, 2004).

Since secondary option can produce by-products such as dirty sludge and waste water which are needed to have further disposal, therefore, this study will focus on how to reduce tar formation by primary measure such as the operating parameters of the gasifier. Consequently, the aim of this research is to investigate the influence of air flow rate on the amount of tar, size of tar compounds and the specified tar compounds produced during gasification.

2. Materials and Methods

2.1 Feedstock

Feedstock used in this study was refuse derived fuel (RDF) made from reclaimed MSW from a dumpsite. Reclaimed MSW was preferred because it has high composition of combustible wastes, low moisture content, and less biodegradable fractions. Combustible wastes sorted from reclaimed waste were cut into small pieces and briquetted into cylindrical form with an average diameter of 1.2 cm and length of 5 cm. Table 3 shows the main properties of RDF used in this study.

2.2 Experimental setup

The gasification tests were conducted in a 10 kg.h^{-1} laboratory scale downdraft gasifier (Figure 1) using air as gasification agent. The gasifier is 2,000 mm high with diameter of 600 mm operating at atmospheric pressure. The gasifier consists of hopper, drying chamber, pyrolysis chamber, reaction (oxidation or reduction) chamber, and ash chamber.

The experimental setup used in this study is shown in Figure 2. The gasifier has an air inlet connected to an air blower. The regulator was installed to control the amount of air supplied to the gasifier. The producer gas exit pipe was connected to the gas-flare where gas was burned to control air pollution resulting from producer gas released into the atmosphere. In addition, the gasifier was connected to an online data logger via eight K-type-thermocouples, installed along the height of the gasifier, for temperature recording during the gasification process.

2.3 Experimental procedure

At the beginning of the experiments 20-30 kg of RDF was fed into the gasifier. The fuel ignition process was facilitated by the air supply at the flow rate of 100 NL.min^{-1}. As the ignition was well developed, the air supply was set to a selected air flow rate to start each experiment. Different air

Table 1. Requirements of producer gas for gas engine and gas turbine (Knoef, 2005).

Application	Gas engine	Gas turbine
Tar (mg.Nm^{-3})	< 50	< 5

Table 2. Amount of tar and particulate from different types of gasifiers.

	Downdraft	Updraft	Circulating Fluidized Bed
Tar (mg.Nm^{-3})	100-150	12,000	2,000

Table 3. Physical and chemical properties of RDF.

Physical properties	
RDF size, diameter x length (cm)	1.2x5
High Heating Value (MJ/kg)	20.6
Low Heating Value (MJ/kg)	20.59
Moisture content (%wt as received)	1.5
Volatile matter (%wt at dry basis)	60.84
Fixed carbon (%wt at dry basis)	1.53
Ash content (%wt at dry basis)	37.63
Chemical properties	
Carbon (%wt at dry basis)	45
Hydrogen (%wt at dry basis)	6.86
Nitrogen (%wt at dry basis)	2.1
Sulfur (%wt at dry basis)	0.19
Oxygen (%wt at dry basis)	8.22

flow rates of 300, 350, 400, 450, and 500 NL.min^{-1} were selected in this study.

At steady state of gasification process, tar was sampled from the exit pipe according to tar and particle sampling procedure, as elaborated in tar protocol (CEN, 2004). Tar sampling setup consists of six impinger bottles connected in series; five of which had approximate 50 ml of isopropanol and the last was empty for drop trapping (Figure 3). Particulates were removed from the gas by filter before tar collection. Rotary evaporator was used to analyze gravimetric tar while Gas Chromatography and Mass Spectrometer (GC-MS) was used to analyze tar composition. Distribution of tar compounds based on size was done by analyzing compounds with a selected number of carbon atoms, namely C_{10}, C_{17}, and C_{19} representing tar compounds with ten, seventeen, and nineteen carbons, respectively. Composition of selected tar compounds were calculated based on the individual intensity proportional to the total intensity of all tar compounds detected by GC and was noted as percentage relative proportion.

3. Results and Discussion

3.1 Temperature profile

The temperature profiles were analyzed based on the average of temperatures recorded by thermocouples at steady state of gasification process. Table 4 shows the average

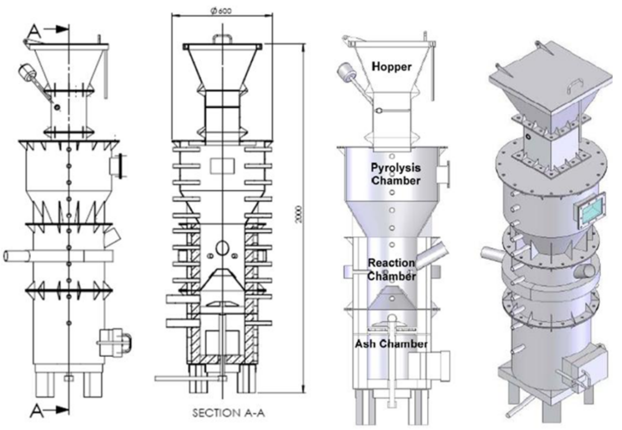


Figure 1. Lab-scale downdraft gasifier.

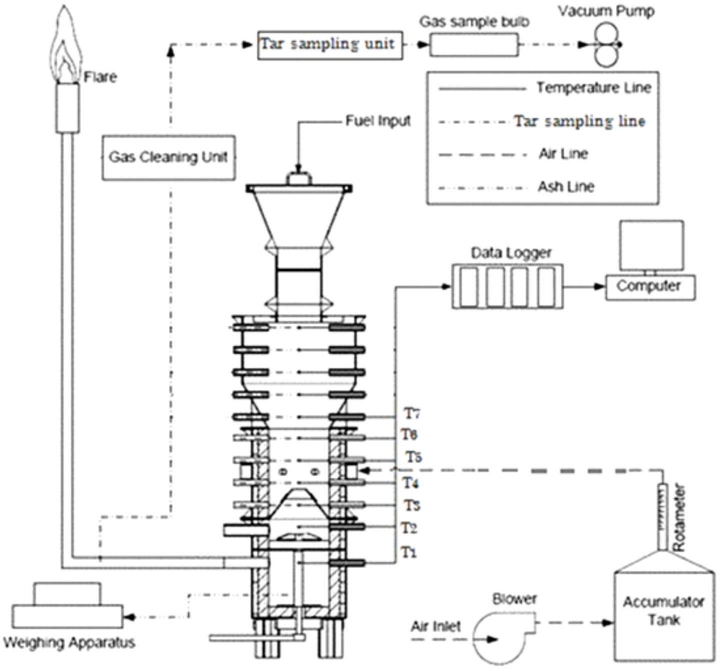


Figure 2. Experimental set up.

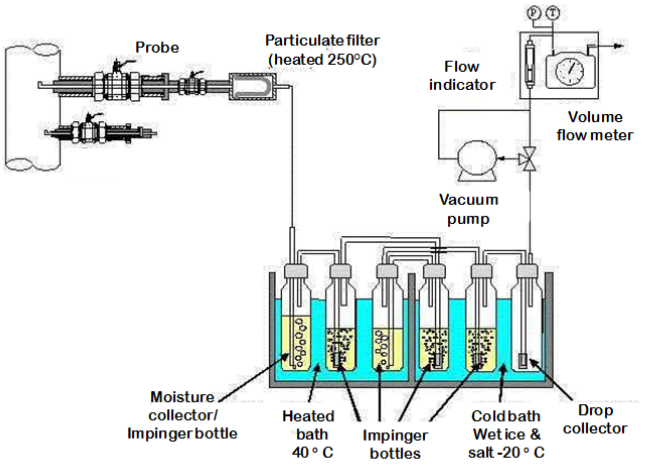


Figure 3. Tar sampling setup adopted from tar protocol (CEN, 2004).

Table 4. Average temperature in different reaction zones during gasification.

Air flow rate (Nl/min)	Temperature (°C)			
	Drying	Pyrolysis	Oxidation	Reduction
300	<200	450	697	505
350	<200	500	719	585
400	<200	500	803	625
450	<200	550	817	685
550	<200	500	870	750

temperature in different reaction zones for all air flow rates. Furthermore, Figure 4 shows temperature profiles along the height of the gasifier for all air flow rates from which locations of reaction zones during gasification, namely drying, pyrolysis, oxidation, and reduction, could be envisaged. For all air flow rates, the drying zone for moisture evaporation occurred near the top of the gasifier (at a height of 58-80 cm) with a temperature below 200°C. Pyrolysis zone was located below drying zone with a temperature of 200-600°C (at a height of 48-58 cm). Oxidation zone took place at a height of 23-48 cm with a temperature of 500-1,000°C. At the bottom of the gasifier between 10 cm and 23 cm, the reduction zone occurred with a temperature of 450-850°C. The temperatures observed are in agreement with literature data (Fagbemi *et al.*, 2001; Knoef, 2005; Kaltschmitt *et al.*, 2009; Verma, 2014).

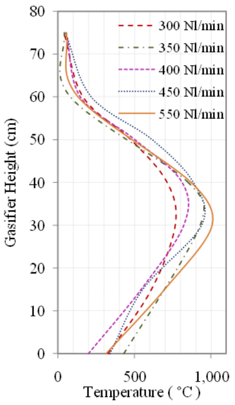


Figure 4. Temperature profiles for various air flow rates.

It was noted that the amount of air supply has direct impact on the temperature inside the gasifier where the temperature increased with increasing air flow rate. This is because the increase in the air flow rate facilitated the exothermic oxidation reaction in the oxidation zone since the amount of fed air was below the stoichiometric requirement.

3.2 Quantity of tar

The gasification was performed using air flow rates of 300-550 NL.min^{-1} in order to investigate the quantities of tars. At the air flow rate of 300 NL.min^{-1} the quantity of tar formed was 15.6 g.Nm^{-3} (Figure 5). A tar reduction rate of 70% was observed with the increase in air flow rate to 550 NL.min^{-1}, in which the tar quantity was 4.7 g.Nm^{-3}. This tar reduction corresponded to an increase in the temperature in the oxidation zone from 697°C to 870°C and an increase in the temperature in the reduction zone from 505°C to 750°C.

Several studies reported tar reduction with the increase in supply of gasifying agent (Turn *et al.*, 1998; Surjosatyo, 2008; Siedlecki and de Jong, 2011; Sarıoğlan, 2012) due to thermal cracking reaction of tar at high temperatures.

3.3 Tar compounds

The results from Figure 6 show the variation of total GC detectable tar compounds with the increase in air flow

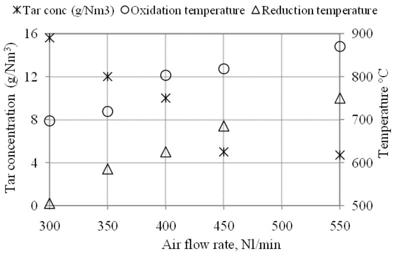


Figure 5. Quantity of tar, oxidation temperature and reduction temperature with air flow rates.

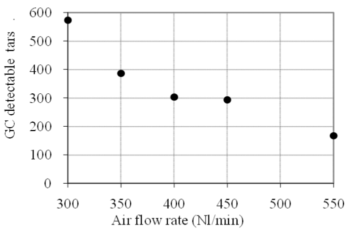


Figure 6. Variation of gas chromatography (GC) detectable tars with air flow rate.

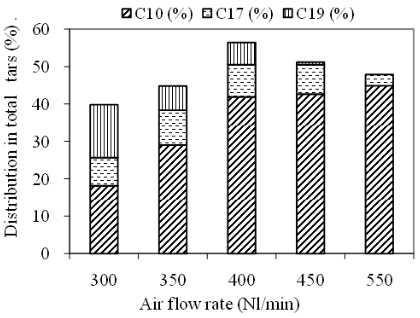


Figure 7. Distribution of C_{10}, C_{17} and C_{19} in the GC detectable tars.

rates. At 300 NL.min^{-1}, GC detected about 573 tar compounds. The increase in air flow rate from 300 to 550 NL.min^{-1} led to a reduction in GC detectable tars to 167 tar compounds showing an overall reduction rate of 70% GC detectable tars.

3.3.1 Tars sizes

Figure 7 indicates the amount of selected tar sizes in the total GC detectable tars. In this study, C_{10}, C_{17} and C_{19} were chosen to represent tar compounds with ten, seventeen and nineteen carbon atoms, respectively. The results showed that the increase in air flow rate led to an increase in the formation of tar species with low carbon number. The amount of C_{10} increased from 18.2% at 300 NL.min^{-1} to 41.9% 400 NL.min^{-1}. A further increase in air flow rate had only a small effect on the amount of the C_{10} compounds. The amount of C_{17} was slightly constant at 7% of GC detectable tars for the air flow rate of 300, 350 and 450 NL.min^{-1}. Increase in air flow rate from 450 NL.min^{-1} to 550 NL.min^{-1} led to reduction in the amount of C_{17} to about 3% of GC detectable tars. C_{19} tar compounds decreased by 50% with an increase in the air flow rate from 300 NL.min^{-1} to 350 NL.min^{-1}. At an air flow rate of 550 NL.min^{-1} a complete removal of C_{19} compounds was observed. These results corresponded to an increase in the temperatures in the reduction zone from 505°C to 750°C. An increase in C_{10} compounds corresponding to a decrease in C_{19} and C_{17} compounds indicates that large tar compounds were cracked into smaller tar compounds at elevated temperature.

Fagbemi observed similar results where tar cracking prevails with the increasing amount of the gasification agent (Fagbemi *et al.*, 2001). In another study, Sarıoğlan (2012) observed the cracking of large molecules into small molecules with respect to the increase in temperature.

3.3.2 Specific tar compounds

As stated by Milne *et al.* (1998), it is important to know specific tar compounds contained in the producer gas in order to estimate their impact on producer gas applications and to optimize gasifier performance. The following tar compounds, phenol, xylene, styrene, phenanthrene, pyrene, indene, toluene, and naphthalene, were selected for the investigation of tar evolution. The selection was based on the important tar compounds present in the producer gas from downdraft gasifiers (Milne *et al.*, 1998; CEN, 2004; Phuphuakrat *et al.*, 2010; Anis and Zainal, 2011).

The results show an increase in toluene, styrene, indene and naphthalene corresponding to the increase in air flow rate from 300 to 550 NL.min^{1}. A sharp increase in toluene was observed when the air flow rate was increased from 450 NL.min^{1} to 550 NL.min^{-1}. A similar trend was noted for toluene and styrene. Furthermore, the increase in air flow rate to from 300 to 350 NL.min^{1} led to large increase of naphthalene, increase in indene and substantial decrease in Pyrene (Figure 8a).

The evolutions of tar compounds with respect to reduction zone temperature are shown in Figure 8b. The results show the dominance in naphthalene at reduction temperatures above 600°C. Indene showed a sharp increase at reduction temperatures above 680°C.

The decrease in pyrene in relation to the increase of naphthalene, toluene and indene shows that the tar cracking was promoted with increasing reduction temperatures. Therefore, pyrene, which is the large tar compound, was cracked to smaller tar compounds, e.g. naphthalene, indene, and toluene. This is in agreement with Sanoglan (2012), whereby large tar molecules (xylene) were cracked into lighter tar molecules (toluene) at higher temperatures. Similar results were reported from other studies, where at high temperature large tar molecules were reduced while light tar molecules dominated the tar composition (Knight, 2000; Phuphuakrat *et al.*, 2010).

3.4 Tar reduction mechanism

The tar reduction can be resulted from several reactions, namely exothermic oxidation reaction (Equation 1), endothermic steam reforming reaction (Equation 2) and endothermic carbon dioxide reforming reaction (Equation 3) (Abu *et al.*, 2004):

$$C_nH_m + \left(\frac{n}{2} + \frac{m}{4}\right)O_2 \rightarrow \mathrm{nCO} + \frac{m}{2}H_2\mathrm{O} + \text{heat} \qquad (1)$$

$$C_nH_m + \mathrm{n}H_2\mathrm{O} + \text{heat} \rightarrow \mathrm{nCO} + \left(\mathrm{n} + \frac{m}{2}\right)H_2 \qquad (2)$$

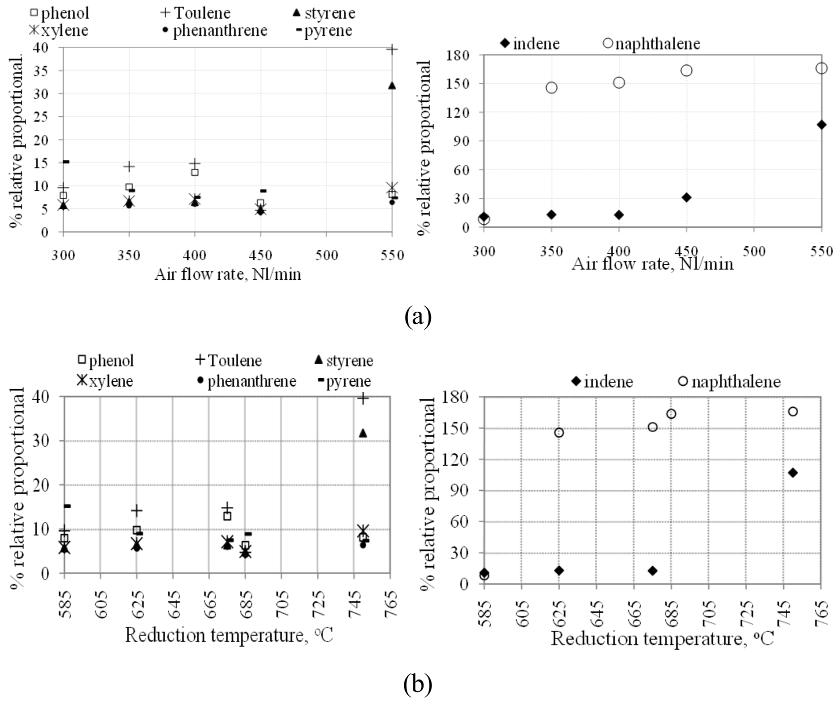
e.g. $\mathrm{C + H_2O \rightarrow CO} \quad \Delta\mathrm{H} = 118.5\mathrm{kJ/mol}$

$$C_nH_m + \mathrm{nCO_2} + \text{heat} \rightarrow \mathrm{2nCO} + \left(\frac{m}{2}\right)H_2 \qquad (3)$$

e.g. $\mathrm{C + CO_2 \rightarrow CO + H_2} \quad \Delta\mathrm{H} = 159.9\mathrm{kJ/mol}$

where C_nH_m is representing tar compounds.

The increase in air flow rate promotes the oxidation reaction (Equation 1) where more oxygen reached oxidation zone and reacted with tars to form carbon monoxide and water vapor, hence tar reaction. In addition, the oxidation reaction is sped up by the increased amount of oxygen; hence,



(a)

(b)

Figure 8. Evolution of tar compounds (a) with air flow rate and (b) with reduction temperature.

high temperatures in the gasifier. The heat released during oxidation reaction is the driving force for the endothermic reactions, presented in Equations 2 and Equation 3, to occur. High temperatures shift the reaction equilibrium to the right, whereby tars are reduced and carbon monoxide and hydrogen are formed.

Additionally, tar reduction can be achieved through cracking or polymerization process. There is a linear relationship between the amount of tar destruction and the amount of cracking or polymerization products. Tar cracking and polymerization depend on the amount of hydrogen atoms present for reactions. Low hydrogen contained in tar prevents the cracking process while it prefers the polymerization process leading to formation of heavy tars. The situation is reversed with high hydrogen content. The hydrogen atom transfer-mechanism observed here is called *'hydrogen shift'*, as shown in Figure 9 (Hoeven, 2007).

In this study, tar was reduced through cracking of large molecules into small molecules. This was clear when analyzing tar sizes (Section 3.3.1). Large tar C_{15} and C_{17} were reduced with the increase in temperature where C_{10} increases. Likewise, reduction of large tar compounds e.g. pyrene (C_{16}) and phenanthrene (C_{14}), corresponded to the increase in naphthalene (C_{10}) and other smaller compounds (Section 3.3.2). In other words, the formation of naphthalene (C_{10}) from pyrene (C_{16}) and phenanthrene (C_{14}) was favored along with its cracking into indene (C_9) and toluene (C_7). Figure 8 shows that even though both naphthalene (C_{10}) and indine (C_9) were formed at higher rates, the rate of formation for naphthalene (C_{10}) was higher than that of indine (C_9). Therefore, naphthalene (C_{10}) is an intermediate compound between tar cracking and formation reactions.

As an example, cracking and polymerization of naphthalene are presented in Figure 10 and Figure 11, respectively. These illustrations are chosen to show the possible tar cracking pathway for this study where naphthalene dominated and indene and toluene increased. In addition, the increase in naphthalene was accompanied by the decrease in large tar molecules e.g. pyrene, indicating the reverse of naphthalene polymerization shown in Figure 11.

4. Conclusions

This study shows that it is possible to reduce the amount of tar formed during gasification by varying one of the important operating parameter of the gasifier that is the air flow rate. The air flow rate, in case of gasification of RDF from reclaimed waste, should be adjusted until the tempera-

$$\text{tar} + H_2 \rightarrow \text{cracking products}$$

Hydrogen shift

$$\text{tar} - H_2 \rightarrow \text{polymerization products}$$

Figure 9. Hydrogen shift.

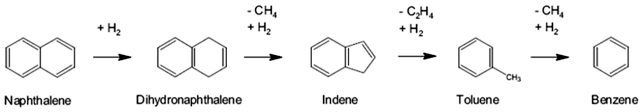


Figure 10. Naphthalene cracking scheme (Hoeven, 2007).

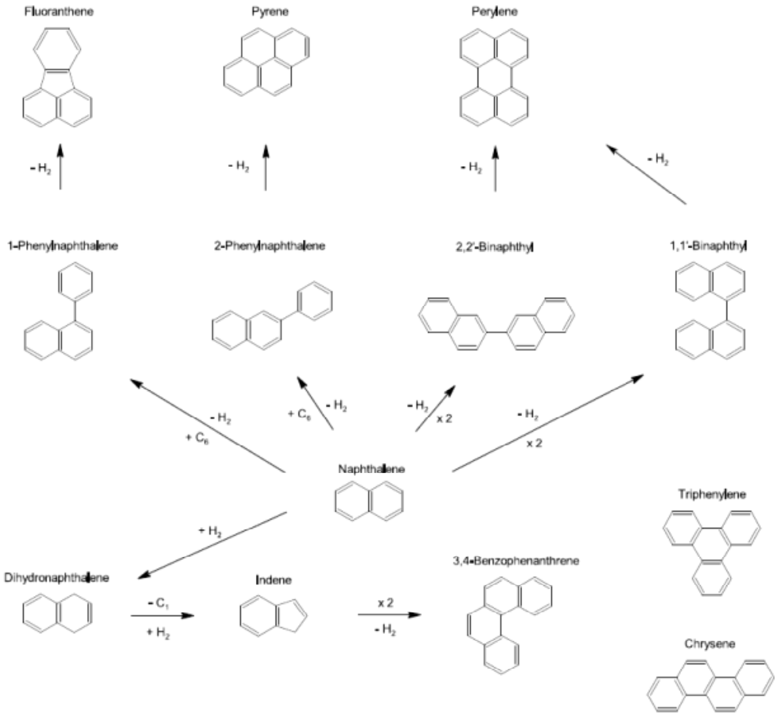


Figure 11. Naphthalene polymerization scheme (Hoeven, 2007).

ture in oxidation zone and reduction zone reaches ≥850°C and ≥750°C, respectively, in order to reduce amount of tar to approximately 5 $g.Nm^{-3}$. In addition, high temperatures facilitate the reduction in the size of tar compounds. The amount and size of tar molecules were reduced with increase in the air flow rate in correlation with the increase in temperatures. Furthermore, high temperatures led to the reduction in heavy tar molecules, while light tar molecules increased due to tar cracking process. However, the amount of tar from this study is still above the minimum tar requirement for gas engine and gas turbine, therefore, the simple second tar removal method is necessary.

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