TAR ANALYSIS IN SYNGAS DERIVED FROM PELLETIZED BIOMASS IN A COMMERCIAL STRATIFIED DOWNDRAFT GASIFIER

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A study was conducted to quantify tar formation in a stratified downdraft gasifier using wood pellets. The effect of biomass flow rate on tar concentration was also analyzed, and more than thirty compounds in tar were quantified. Among the different compounds in tar, tertiary condensed products such as toluene, o/p-xylene, naphthalene, phenol, styrene, and indene were observed in significant amounts. Tar concentration in the syngas was found to be in the range of 340 to 680 mg/Nm^3. These concentrations were found to be much higher when compared to a similar gasifier using woodchips.

Keywords: Downdraft gasifier; Gasification; Pellets; Syngas; Tar

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INTRODUCTION

The biomass gasification process can be used for “green” power or fuel production. It is a relatively mature technology compared to other thermochemical and biochemical processes. Nonetheless, tar concentration in syngas is one of the major hurdles standing in the way of syngas utilization. Milne et al. (1998) defined tar from the gasification process as a material in the syngas that condenses inside a gasifier or in the equipment used for transporting the product stream to its end use. Tar compounds are largely aromatic in nature and can be classified into four groups, which are shown in Table 1.

Table 1. Classification of Tar from Thermal Cracking of Biomass

Classification	Tar compounds
Primary	cellulose-derived products such as levoglucosan, hydroxyacetaldehyde, and furfurals and similar hemicelluloses and lignin-derived products
Secondary	phenolics and olefins
Alkyl tertiary	methyl derivatives of aromatics
Condensed tertiary	benzene, naphthalene,acenaphthylene, anthracene/phenanthrene, pyrene

Among the different types of products listed in Table 1, condensed tertiary products are formed as a result of consecutive conversion of primary tar at high temperature. Therefore, these two types of products, condensed tertiary and primary tar products, are not usually found in the syngas at the same time (Milne et al. 1998). The maximum limit of tar concentration in syngas varies depending upon its end use. The tolerable limit of tar concentration in syngas is 500 mg/Nm^3 (normal cubic meter), 100 mg/Nm^3, 0.5 mg/Nm^3, and 5 mg/Nm^3 for compressors, internal combustion engines, methanol synthesis, and gas turbines, respectively (Milne et al. 1998). Tar production in a downdraft gasifier is much lower than in both updraft and fluidized-bed gasifiers. However, the process may not meet the requirements needed to be used directly without prior treatment in power generation applications and liquid fuel synthesis processes (Warnecke 2000).

Liquid fuel synthesis from syngas requires high purity in the reacting gases; thus the tar must be removed. The major problem with tar, when used in power generation, is condensation at low temperature, which creates blocking as well as fouling in power plant equipment such as economizers and air-preheaters (Devi et al. 2003). Hence, subsequent treatment is usually warranted depending upon the end use of the syngas. Also, the nature of tar from gasification varies according to its design.

Downdraft gasifiers produce tertiary tar, while tar from updraft gasifiers contain mostly primary tar due to a lower potential of tar cracking inside the gasifier (Milne et al. 1998). Syngas from fluidized-bed gasifiers contain tar, which is a mixture of secondary and tertiary tar (Milne et al. 1998). Tar content in a downdraft gasifier is usually in the range of 0.01 to 6 g/Nm^3, while updraft and fluidized-bed gasifiers usually have an average tar content of 50 g/Nm^3 and 6 to 12 g/Nm^3, respectively (Milne et al. 1998). Residence time, oxidizing agents (steam versus air or oxygen), and temperature in the gasification and reduction zones are the most important factors in determining the level of tar in a downdraft gasifier (Monteiro Nunes et al. 2007). As temperature increases, tar content in the syngas decreases due to thermal cracking (Han and Kim 2008). Li et al. (2004) have reported that with an increase in temperature from about 700^{o}C to 820^{o}C, tar content decreases significantly from 15 to 0.54 g/Nm^3 in a circulating fluidized-bed gasifier.

Figure 1 shows the relationship between gasifier reaction temperature and tar yield (Baker et al. 1988). It can be observed from the figure that as temperature increases, the yield of condensable liquids (tar) is significantly reduced. Increase in equivalence ratio also decreases tar content at the expense of higher levels of combustion inside the gasifier. This results in a higher concentration of CO_2, which is an undesirable product (Lv et al. 2004).

Although tar concentration in syngas from a downdraft gasifier is usually lower, these tars are also more stable and might be difficult to crack and remove depending upon the end-need (Beenackers 1999). For use in an internal combustion engine, concentration of tar should be less than 100 mg/Nm^3 for successful long-term operation (Hasler and Nussbaumer 1999 Milne et al. 1998).

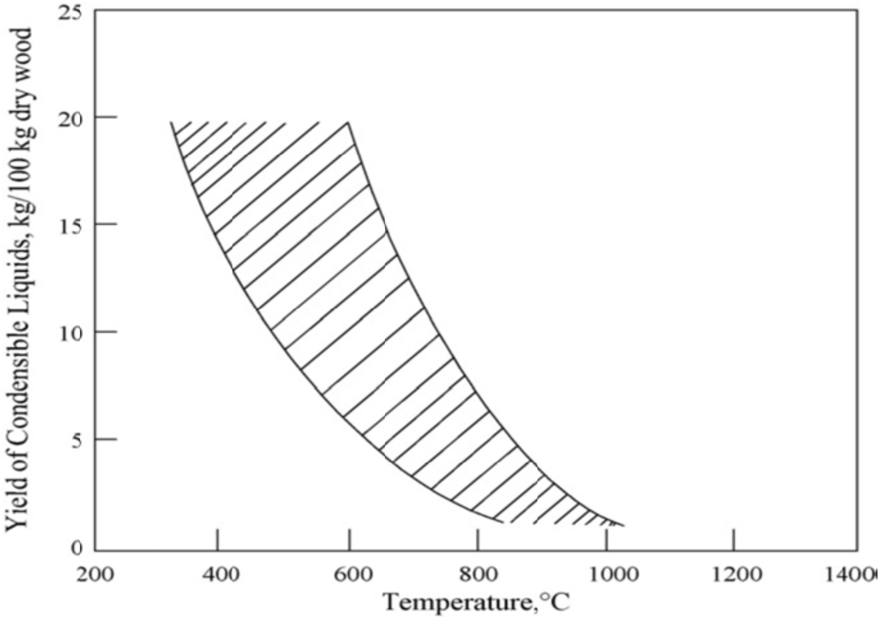

Fig. 1. Effect of maximum reactor temperature on tar production (adopted from Baker et al., 1988, pp. 1-11)

Milne et al. (1998) have discussed the tar reduction procedure as any one of physical, thermal, or catalytic techniques. Han and Kim (2008) have classified tar reduction methods into five groups: mechanism method, self-modification, thermal-cracking, catalytic cracking, and plasma method. A system that includes a scrubber, cyclone, and electrostatic precipitator can effectively remove 40 to 99% of tar in syngas, but the useful energy that can be achieved from tar conversion is lost. In other methods, tar is converted into other gases, and this increases the heating value of the syngas, thus increasing the energetic efficiency of the process. Devi et al. (2003) suggested three methods for tar removal: adjustments of the operational parameters, addition of bed additives/catalysts, and gasifier modification.

It was observed from a few trial studies that more tar was accumulated in filters while running pelletized biomass compared to pine wood chips in a CPC (Community Power Corporation) gasifier. Therefore, the objective of this study was to characterize and quantify tar from the CPC gasifier using pelletized biomass, and to compare experimental results with the literature.

EXPERIMENTAL

Experimental Set-Up and Procedure

All of the gasification experiments were carried out in a commercial-scale stratified downdraft gasifier (25 kW_e) developed by the Community Power Corporation (CPC, Littleton, CO). A photograph of the gasifier is shown in Fig. 2. A known amount of biomass was fed into the gasifier, and the run-time was noted in order to calculate the average biomass feed rate. Biomass feed rate was controlled based on a specified level of biomass inside the gasifier. A level sensor detected the level of biomass in the gasifier and turned the feeder (auger) once the biomass level fell below the set value. Air was used as an oxidizing agent for biomass gasification. Primary air in the gasifier was obtained from the open top of the gasifier. The gasifier had multiple secondary air injection nozzles (tubes around the gasifier) where the secondary air was fed with a 250 W (1/3 HP) air blower. The purpose of the secondary air supply was to improve the combustion reaction, and also to maintain uniformity in temperature along the length of the gasifier. The grate was shaken at an adjustable regular interval via a grate-shaker mechanism to remove the ash formed during the operation. The gasifier was also shaken at a regular interval to facilitate the smooth flow of biomass inside the gasifier and to prevent channeling and bridging inside the gasifier. Charcoal left from the previous experiment (or fresh charcoal for first experiment) inside the gasifier was ignited with an igniter before the fresh biomass was fed. The height and inside diameter of the gasifier reactor were 1200 mm and 350 mm, respectively. Data were collected once the gasifier reached steady state. The steady state of the system was indicated by having constant temperature across the different levels of the gasifier, most commonly 800˚C at any three locations among T1-T4. The time required to attain steady-state generally varied from 30 min to 1 hr and was affected by biomass type and outside weather conditions.

Commercial wood pellets were obtained from American Wood Fiber (Columbia, MD). Wood pellets were fed into the gasifier by an external biomass feeder to provide an accurate measurement of the mass used in each experiment. Immediately after the gasifier, syngas was sampled. The experiments with commercial wood pellets (once steady state was achieved) were run for almost 4 hours. The gasifier was not designed to vary biomass feed rates; therefore, an alternative approach was adopted to obtain different biomass flow rates. Since the syngas output rate depends on the biomass feed rate into the system, the syngas output rate (which can be easily controlled in this gasifier) was varied to control the biomass feed rate. Results from ultimate and proximate analyses of wood pellets used for these experiments are shown in Table 2.

Table 2. Characterization of Wood Pellets used for Gasification Study[‡]

Sample	**Wood pellets**
Carbon, wt.%	47.90
Hydrogen, wt.%	6.02
Nitrogen, wt.%	0.04
Oxygen[†], wt.%	45.60
Ash content, wt.%	0.44
Higher heating value (MJ/kg)	18.34

[‡] dry basis; [†] calculated from difference.

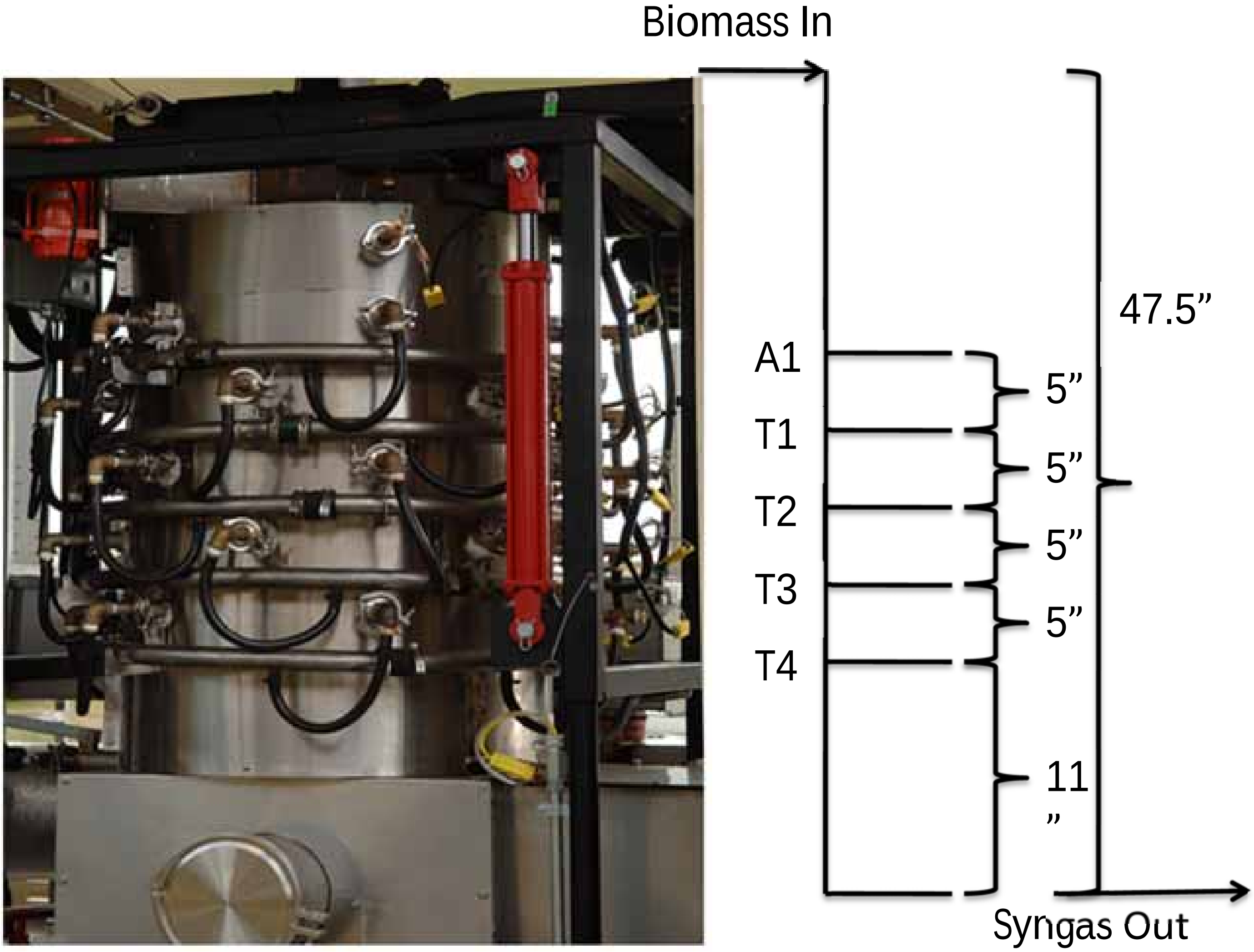

Fig. 2. Photograph of the CPC downdraft gasifier (All dimensions are in inches and not to scale)

Figure 3 shows a schematic diagram of the experimental set-up used to measure tar concentration in syngas from the gasification of wood pellets. Syngas was sampled from the port immediately after the downdraft gasifier, and is then passed through impinger bottles (each containing 50 mL of isopropyl alcohol). The first impinger bottle was kept at ambient conditions while the other two were kept in an ice-bath in order to maintain a temperature around the freezing point of water. The tar present in the syngas condenses under these conditions in the impinger bottles and can later be quantified. The water absorber, used after the impinger bottles, attracts all the moisture present in the syngas stream after condensation, leaving it dry. A flow-meter placed after the water-absorber measures the syngas flow rate that is required to find the tar concentration per standard volume.

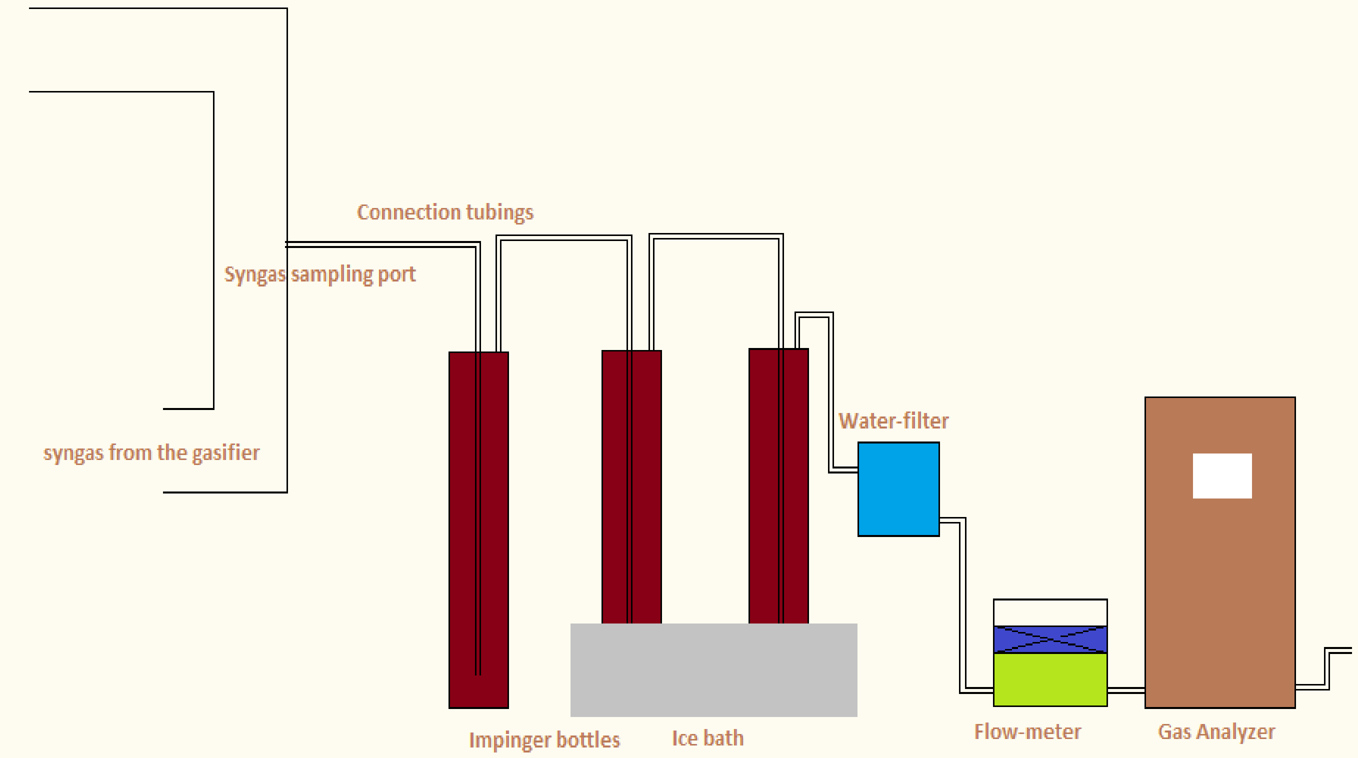

Fig. 3. Experimental set-up for tar quantification

The tar components were analyzed with an Agilent 7890 GC/5975MS using a DB-1701 column (30 m; 0.25 mm i.d.; 0.25 mm film thickness). Thirty-one compounds were selected for quantification, and five data points were generated in such a way that concentration of tar compounds fell within those five points. The tar, which had been previously dissolved in isopropyl alcohol, was further diluted five times with dichloromethane. A dilute tar sample was injected into the column. Each sample was injected twice. Splitless injection was selected. The injector and the GC/MS interface were kept at constant temperatures of 280°C and 250°C, respectively. The initial temperature of the column, 40°C, was maintained for 2 min, the temperature was subsequently increased to 250°C at 5°C/min, and the final temperature was held for 8 min. Helium of ultra-high purity (99.999%) was used as a carrier gas and flowed at 1.25 mL/min. Compounds were ionized at 70 eV electron impact conditions and analyzed over a mass per change (m/z) range of 50 – 550. Tar compounds were identified by comparing the mass spectra with the NIST (National Institute of Standards and Technology) mass spectral library.

RESULTS AND DISCUSSION

Tar Concentration in Syngas

Table 4 shows the various tar compounds along with the amount obtained from the test runs from the gasifier. The major constituents observed in tar are similar to those observed by other studies.

Table 4. Quantification of Tar Constituents in Syngas from Experiments

Tar Compounds	**Concentration (mg/Nm^3)**
Toluene	76.8-198.3
o/p-Xylene	9.3-111.6
Naphthalene	62.3-126.1
Phenol	6.9-67.2
Styrene	21.0-65.1
Indene	15.7-55.8
Ethylbenzene	2.5-25.0
Phenol, 3-methyl-	1.3-25.4
Benzofuran	8.5-24.9
Biphenylene	7.1-22.2
Benzofuran, 2-methyl-	0-23.8
Benzene, 1-ethenyl-3-methyl-; (m-Methylstyrene)	6.6-18.8
Naphthalene, 2-methyl-	5.1-16.2
Naphthalene, 1-methyl-	5.9-14.6
Biphenyl	2.6-10.1
Phenol, 2-methyl-	0.5-8.9
Naphthalene, 2-ethenyl-; (2-Vinylnaphthalene)	0.4-6.7
Furfural	0-4.0
Naphthalene, 1,8-dimethyl-	0.6-3.6
Naphthalene, 1,5-dimethyl-	0-3.6
Dibenzofuran	0.4-3.4
.alpha.-Methylstyrene	1.5-3.1
Benzene, 1-ethyl-2-methyl-; (2-Ethyltoluene)	0.6-3.0
Benzene, 1,2,3-trimethyl-	1.4-2.4
Phenol, 2,4-dimethyl-	0-2.4
Acenaphthene	0.3-2.1
Phenol, 3,5-dimethyl-	0-1.9
Naphthalene, 2,3-dimethyl-	0-1.4
Phenol, 3-ethyl-	0-1.3
Phenol, 4-ethyl-	0-1.0
Naphthalene, 1,8-dimethyl-	0-0.8
Total	340-680

Bari et al. (2000) reported toluene, ethylbenzene, styrene, and p-xylene as major tar constituents in the syngas obtained from the gasification of feedstocks such as almond shells and oak in a downdraft gasifier using air as a gasifying medium. Similar results were reported by Yamazaki et al. (2005) in an experimental investigation of the effect of superficial velocity on tar concentration in a downdraft gasifier using fir wood chips as a feedstock. As expected, the majority of tar compounds observed in higher proportions are tertiary condensed tar products due to thermal cracking inside the gasifier. Figure 4 shows the fraction of various compounds in tar based on averages of 13 experiments.

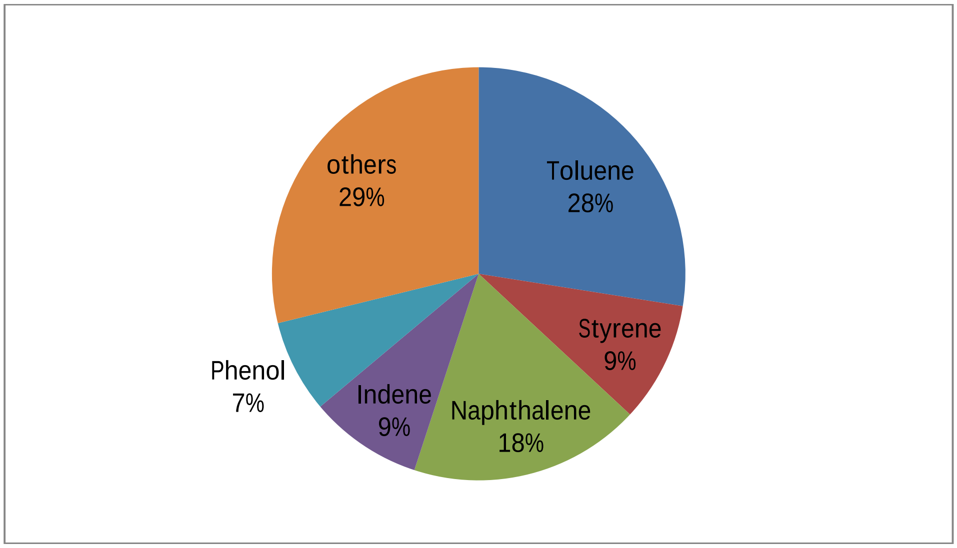

Fig. 4. Distribution of different tar compounds in syngas from a downdraft gasifier

Figure 5 shows the effect of biomass flow rate on total tar concentration in a stratified downdraft gasifier. The moisture content of wood pellets was between 2.7 and 5.3 wt.% (wet basis), and the equivalence ratio was in the range of 0.28 to 0.37. Other conditions were kept constant for all the experiments. Tar concentration in syngas from this stratified downdraft gasifier was found to be 340 to 680 mg/Nm^3. Further, moisture contents and equivalence ratios (not shown here) did not show any trend for tar concentrations for the CPC gasifier.

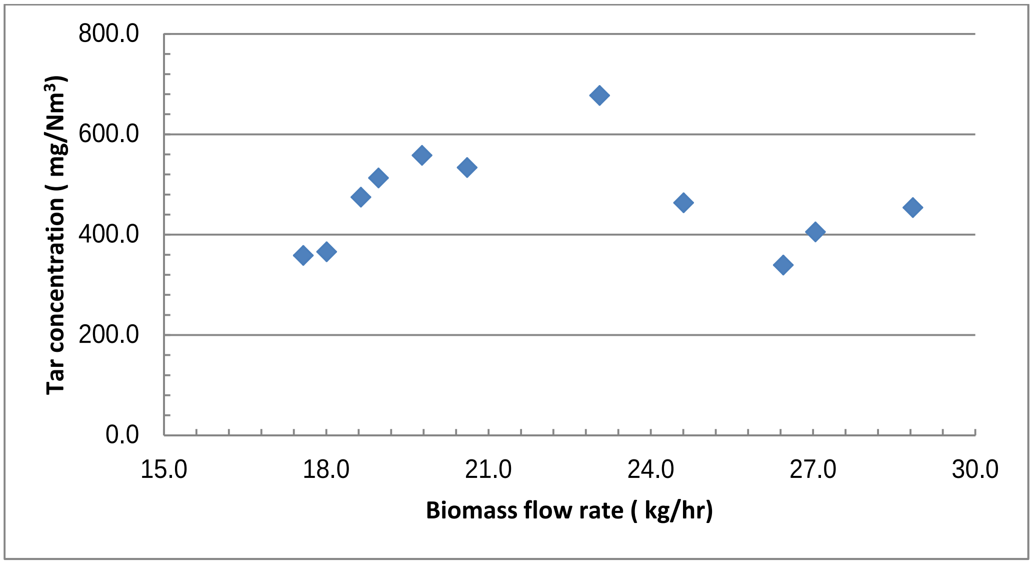

Fig. 5. Effect of biomass flow rate on tar concentration

Dogru et al. (2002) and Phuphukrat et al. (2010) reported tar concentrations of 6.37 and 8.38 g/Nm^3 for throated and throat-less downdraft gasifiers, respectively, while using sewage sludge as a feedstock. In another study conducted in a similar type of downdraft gasifier (also purchased from CPC), Wei (2005) reported a tar concentration of 54 mg/Nm^3 when using wood chips as a feedstock, which is significantly lower than that of this study. This might be due to the difference in bulk density of wood pellets and wood chips. Since wood pellets are more than three times as dense as wood chips, the temperature at the core of wood pellets might be lower than that at the surface, producing higher tar concentration when compared to wood chips. This is an interesting finding and if the quality of syngas is important in downstream processing, the size of the feedstock plays an important role and must be optimized.

CONCLUSIONS

1. Tar concentration from the downdraft gasifier was mostly comprised of condensed tertiary products with a significant amount of toluene (76.8-198.3 mg/Nm^3), o/p-xylene (9.3-11.6 mg/Nm^3), naphthalene (62.3-126.1 mg/Nm^3), phenol (6.9-67.2 mg/Nm^3), styrene (21-65.1 mg/Nm^3), and indene (15.7-55.8 mg/Nm^3). This shows that primary and secondary tar cracking is very efficient in the current configuration of the downdraft gasifier.
2. Tar concentration was also significantly lower than those reported by others in similar studies with conventional downdraft gasifiers. However, the tar concentration from the gasification of pellets was found to be significantly higher than those with the gasification of wood chips in a similar type of gasifier.

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