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A Wet Packed-bed Scrubber for Removing Tar from Biomass Producer Gas

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Abstract

In this work, we report the performance of a lab scale wet packed-bed scrubber using woodchips as a packing material. Wood chips were wetted by waste cooking oil which acted as a scrubber medium. Residence time of 8.5 s was comparable to the packed-bed filter used in a pilot scale gasifier. We studied the effects of oil/gas ratio (4.7 and7 $m^3/s/m^3/s \times 10^3$), oil temperature (ambient, ~23 °C and 50 °C) and woodchips size (coarse= ~0.02-0.045 m, 0.003-0.03 m and 0.002-0.007 m and fine= (0.015-0.04 m) × (0.002-0.005 m) × (0.001-0.003 m)) to remove tar model compounds from producer gas. The results demonstrate liquid to gas ratio has significant effect (p-value= 0.002) on removing toluene as tar model compound; next important parameter is bed configuration (p-value= 0.014), impacting toluene removal efficiency. For naphthalene, oil flow rate (p-value= 0.022) and bed configuration (p-value= 0.024) have almost similar effects. Oil temperature has relatively lower impact on removing tar.

Keywords: Gasification, Tar removal, Wet packed-bed, Woodchips, Waste cooking oil, Naphthalene, Toluene

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1. Introduction

Biomass gasification produces syngas such as CO, H_2 and CH_4 that are desired composition. In addition to syngas, contaminants like particulates, tar and char are formed. Particulates cause erosion of metallic component, alkali metals cause metal corrosion at high temperature, fuel-bonded nitrogen forms NO_x during combustion, sulfur and chlorine could produce dangerous pollutants and acid corrosion of metals (Anis and Zainal, 2011). A wide spectrum of aromatic hydrocarbons containing single and multiple ring aromatics are formed during the gasification, referred to as tar. Tar might plug and foul downstream components (Tarnpradab et al., 2017) and produce metallic corrosion (Anis and Zainal, 2011). Typical tar dew points are between 150 °C and 350°C, which is usually far above the lowest process temperature (∼30°C). Tar along with producer gas condenses and polymerizes inside the process piping, plugs filters and forms damaging deposits inside the engine (Woolcock and Brown, 2013). Therefore it is important to reduce the tar concentration and keep the temperature above the dew point of the tar components. The main barrier in commercialization of small-scale, biomass gasification combined heat and power (CHP) technology is lack of a cost-effective technology to remove tar from the producer gas.

Tar components are categorized into five classes based on their chemical structure, solubility and condensability. Gas Chromatograph-undetectable (relatively higher molecular weight compounds) tar is removed easily at higher temperature and light aromatics (1 ring) are acceptable in engine fuels. Heterocyclic tars that are highly soluble in water, heavy poly-aromatic hydrocarbon (PAH) compounds (4–7 rings) that condense at high-temperatures (>270˚C) even at low concentrations, and light PAH compounds (2–3 rings) that condense at low temperature even at very low concentration are difficult to manage (Anis and Zainal, 2011). In many producer gas applications the desired concentration of tar and particulates are ≤50 mg/m^3 and ≤ 5 mg/m^3, respectively (Table 1).

Table 1-Tar tolerance limit at different applications, ((Pathak et al., 2007)**, Thapa et al., 2017).**

Application	**Tar tolerance limit (mg/Nm^3)**
Compressors	50–500
Internal combustion engines	50–100
Direct fired gas turbines	5
Methanol synthesis	0.1

Tar removal methods such as catalytic cracking, thermal cracking and plasma gasification are suitable for large-scale systems due to their high reliability and efficiency. For small-scale applications, gas clean-up preferably comprises of using tar condenser-scrubbers, packed bed filters and cyclones due to their low cost and relative simplicity (Tarnpradab et al., 2017). Combination of oil scrubber and char-bed filter can eliminate up to 98% tar (Nakamura et al., 2016). In physical tar removal methods, hybrid systems are more efficient than single step methods. Addition of heat exchanger and canola oil scrubber before wood shaving filter increased tar removal efficiency to 61.5% and 97.5% from 10.5% (Thapa et al., 2017). A wet packed-bed constituted of two layers of 15 mm and 6 mm of raschig ring and a layer of stone and water as scrubber medium is able to remove 75% of tar + particulates when their initial concentration is less than 600 mg/m^3 (Bhave et al., 2008). OLGA gas cleaning technology that comprises two scrubbers and a stripper and is built based on the tar dew point is able to remove 99% of phenolic compounds and 95% of heterocyclic tar compounds (class 2) (Boerrigter et al., 2005). Scrubbers, filters and cyclones require less energy compared to catalytic and thermal cracking processes (Bhoi et al., 2015b). Low pressure operation and relative ease of fabrication reduce capital and operational costs of scrubbers. A wet scrubber is able to collect fine particles (<0.3μm) as well (Bhave et al., 2008). The collected tar, oil scrubber (Thapa et al., 2017) and woodchips can be mixed and fed to the gasifier (Tarnpradab et al., 2017).

Scrubbing media should have high efficiency in removing tar and a longer lifetime (Tarnpradab et al., 2017). Water is the most common existing scrubber medium, however, a large number of organics that constitute tar have low solubility in water (Phuphuakrat et al., 2011). Some tar components (e.g. benzene, toluene, xylene, styrene, and indene) dissolve in non-polar adsorbents like vegetable oil and some (e.g. phenols) dissolve in polar solvents such as water. Oily chemicals remove phenol through van der Waals forces. Petroleum diesel, biodiesel, vegetable oil and engine oil that are able to absorb volatile organic compounds (VOCs) have potential to remove tar from biomass producer gas. Vegetable oil (60.1%) is more efficient than water (31.8%) and engine oil (34.6%) in removing tar. Using diesel and biodiesel increase the tar concentration 144.1% and 54.2%, respectively. Biodiesel and diesel evaporate slowly at much lower temperature than their boiling points and increase gravimetric tar concentration of the producer gas. Petroleum diesel and biodiesel boiling ranges are 160–371 ˚C and 200–347 ˚C, respectively (Phuphuakrat et al., 2011). Oils can have a high removal efficiency. For example, sunflower oil as a scrubber medium removes 98% of tar or waste palm cooking oil is able to reduce tar content to as low as 0.022 g/Nm^3 (Thapa et al., 2017). Physical properties of oil, especially its viscosity, affects absorption efficiency (Phuphuakrat et al., 2011). Mass transfer increases at lower viscosity (Bhoi et al., 2015b). The lower the viscosity of vegetable oils, the higher the mass transfer of volatile organic compounds (VOCs) due to decrease in thickness of interface layer on the liquid side that enhances the diffusion process (Bhoi, 2014; Heymes et al., 2006). Light tar removal improves by using lower viscosity scrubber media. However, biodiesel that has a higher viscosity compared to diesel removes light tars better than diesel (Phuphuakrat et al., 2011) perhaps due to higher solubility of tar components in esters. Canola seed oil is able to remove 76.6% of gravimetric tar and increased to 87.6% using 7.5% emulsified absorbent. Light tar removal was similar using pure oil or 7.5% emulsified absorbent (Unyaphan et al., 2016). Wet scrubbers may eliminate certain chlorine compounds, especially hydrochloric acid (HCl)(Balas et al., 2014). Beside the type of solvent, operating temperature affects the solubility of tar with different chemical components (Bassil et al., 2017). Increasing the bio-oil temperature from 40˚C to 60˚C reduced the tar removal efficiency from 63.6% to 51.6% (Nakamura et al., 2016). The effect of temperature on removing tar from producer gas depends on the scrubber media, as well. Increasing the water temperature as a scrubber medium from 5 ˚C to 35 ˚C reduced removal efficiency from 90% to ~60%; while, increasing the temperature even up to 80˚C has no effect on removal efficiency when rapeseed oil methyl ester was used as a scrubber medium (Balas et al., 2014). So, oil give us the chance to work at higher temperature. Overall, vegetable oils are promising solvents to remove tars owing to characteristics of high absorption capacity for tar compounds, availability in large quantities, lower volatility, no health or explosion hazards, and low cost (Bhoi, 2014). Also, these type of solvents are plant-based and CO_2 neutral. Their volatility is lower compared to chemical solvents or even biodiesel and lubricating oil (Bhoi et al., 2015b).

Use of packings in wet scrubbers increases mass transfer due to larger contact area in comparable volumes. Tar and water vapours contact with packing surface and diffuse in liquid from gas (Bhave et al., 2008). There are different types, sizes and materials for packing. Tar removal and liquid distribution improve at lower size of packing (Bhoi, 2014). Packing to column diameter of less than one eighth minimizes the liquid maldistribution (Bhoi et al., 2015a). The bed height should be optimized by considering the parameters such as material cost and pressure drop. Increasing the packed-bed height will increase the contact time and mass transfer area and consequently tar removal efficiency , but, reduces the mass transfer gradient. At low solvent temperature and high scrubber medium flow rate, the effect of bed height is not considerable due to the higher driving force and consequently absorption efficiency (Bhoi et al., 2015a).

In designing a packed bed reactor the important parameters are type, material and size of the packing material, the height of packed-bed reactor, packing method and liquid distribution. Produced gas and liquid fluxes, liquid viscosity, concentration of absorbent in the scrubber media and pollutant concentration affect the absorption system efficiency. Liquid flow pattern and packing wettability depend on the liquid viscosity. Ratio of solvent to tar (w/w), contact time and solvent temperature affect tar removal efficiency (Bhoi et al., 2015c).

Gas and liquid velocity, and specific surface area affect the tar removal efficiency. The mass transfer coefficient depends on gas and liquid viscosity, density and consequently on temperature. Vapour-liquid equilibrium ratio (K-value) demonstrates the mole fraction ratio of a species in two phases at equilibrium and is an important parameter in designing an adsorption column. Liquid and gas flux ratio (L/G) is one of the important design parameters to achieve the desired tar removal efficiency. The minimum L/G ratio can be considered equal to K-value. But the actual liquid flow rate should be 20 to 50% higher than the calculated rate. At equilibrium condition, absorption efficiency reaches to zero. K-value of toluene is 0.1 (Bhoi et al., 2015a). Column diameter is limited by flooding and pressure drop. Gas velocity should be 50% to 70% of the flooding velocity and pressure drop should be less than 1225.7 Pa/m of packed bed (Bhoi et al., 2015a). Scrubbing media surface tension (σ) affects wettability and consequently mass transfer efficiencies (Bhoi et al., 2015a). Mass transfer reduces due to poor wettability at a surface tension of higher than 70 mN/m. Surface tension of lower than 25mN/m is recommended. Surface tension of vegetable oils are approximately 34 mN/m at a temperature range of 30 to 50˚C (Bhoi, 2014) , which is an acceptable range. Lower k-value demonstrates higher removal efficiency because of higher driving force to remove tar from gas phase. At lower temperature, viscosity and density of vegetable oil decreases that increase interfacial mass transfer area and consequently tar removal efficiency. Also, wettability increases at lower temperature because of surface tension reduction. Toluene, benzene and ethylbenzene removal efficiency reduced ~45%, 30% and 35%, respectively by increasing the temperature of oils such as soya oil and canola oil from 30˚C to 50˚C (Bhoi et al., 2015b). Increasing the solvent flow rate will improve the tar removal efficiency by increasing the driving force, but it should be optimized because of its effect on gas velocity and gas-liquid contact time. Enhancement of liquid flow rate will augment the liquid film thickness and liquid hold up. Increase of liquid film thickness increments the mass transfer resistance in liquid phase. Growth of liquid hold up reduces column cross sectional area and consequently reduces residence time by increasing the gas velocity (Bhoi et al., 2015b).

In this work, we fabricated a wet packed-bed scrubber similar to the lab scale setup that Bhoi et al, 2015 (Bhoi et al., 2015a) designed to observe the effect of cooking oil on removing tar. We used woodchips as packing rather than stainless steel. A challenge in using a packing is regular chemical consumption for removing tar from the packing (Bhave et al., 2008). Using woodchips as a packing material eliminate the drawback of using chemicals to clean the contaminated packings regularly. The remaining waste cooking oil on woodchips and tar contaminated oil can be used as a fuel and gasify in the system. We study the effect of oil/gas ratio (L/G), oil temperature, and packing size on tar removal efficiency. We kept the packed-bed height constant at 65 cm in all experiments to provide the same residence time for producer gas as a current packed-bed filter that we are using to clean the producer gas generated in a pilot plant gasifier.

2. Materials and methods

2.1 Scrubber media and tar model compound

Toluene and is propylene were obtained from Sigma-Aldrich, naphthalene from Sigma and anthracene from Aldrich Chemical Company, Inc. Waste cooking oil was kindly donated by a close-by restaurant. We removed the residual food from waste cooking oil by passing it through a filter paper, quantitative 474, cat no. 28306-368. GC-MS determined oleic and octadecanoic acids were major components of cooking oil. Oleic acid is a monounsaturated fatty acid whereas stearic acid by IUPAC name of octadecanoic acid is a saturated fatty acid. Both stearic and oleic acids are lipophilic in nature and can mix well with tar compound such as toluene that are lipophilic (Bhoi et al., 2015a).

2.2 Experimental

Figure 1 demonstrates the schematic of the setup we used to perform the experiments. Gas flow rate (13 L/min), tar (1.5 g/m^3) and humidity concentration (15%) and residence time (8.5s) represent the pilot-plant gasifier that is currently operational. To prepare a model tar, we mixed toluene (84.8%-1 ring), naphthalene (15%-2 rings) and anthracene (0.2%-3 rings) that represent one, two and three ring compounds and constitute 87% of tar. Typical tar composition contains one-ring (benzene (37.9), toluene (14.3), other(13.9)), two rings (naphthalene(9.6), other (7.8)) and three rings (3.6%) 87% of typical tar (Singh et al., 2005)

To simulate the producer gas, first, we mixed nitrogen with water and tar. A cole-parmer gas flow controller (±0.2 L/min) adjusted the nitrogen to $2x10^{-4}$ m^3/s (12 L/min). N_2 heated up to 160 ˚C with a heating tape that wrapped around the tube and its temperature controlled with a variac autotransformer type W5MT3. To prevent droplet formation and consequently sudden decrease of gas temperature we sprayed water (Figure 2). To do so, another cole-parmer gas flow controller (±0.1 L/min) introduced 1.67x10-5 m^3/s (1 L/min) N2 into a tee joint, a MasterFlex peristaltic pump from cole-parmer (±0.1% speed control accuracy) introduced 2 ml/min water into the joint. N_2 pushed water and the mixture of water and gas passed through a <1x10^{-4} m nozzle. This configuration reduced gas temperature fluctuation considerably (Figure 2). A Harvard apparatus syringe pump (PHD/ULTRA), flow rates are accurate within 0.25% and reproducibility within 0.05%, injected 2.3x10-10 m^3/s (13.8 µl/min) tar into the hot moistened gas to reach the desired concentration of tar compound (~0.828 g/m^3). The gas was introduced to a wet packed-bed scrubber. The wet packed-bed scrubber was a stainless steel column with internal diameter of 0.0508m. A sump was connected to the bottom of the column with a flange. A pump supplied the desired pressure (<1x10^{5}Pa) to circulate the oil and its flow was regulated by a liquid flow controller. Oil was sprayed from the top of the column. To deliver the oil efficiently we installed a distributor at 0.05m from the top of the packed-bed. This distributor had 5 holes of 0.0015m diameter which provided 1770 drip point/m^2. In large scale columns, the ideal drip points to provide suitable liquid distribution and consequently efficient mass transfer would be between 60 to 100 points per m^2. A combination of a heater and a fan adjusted desired oil temperature through a control box. In order to reduce the heat losses, a 0.02 m thick fiberglass sheet insulated the column. The gas and oil were contacted in counter-current mode. A stainless steel filter (~0.02m) above the gas entrance kept the woodchip packing media in place. Two traps before and after the column were used for gas sampling. Part of the gas at a flow rate of 1.67x10^{-6} m^3/s (100 ml/min) was continuously passed through a liquid trap filled with 1x10^{-4}m^3 (100ml) of isopropanol using a Masterflex L/S Digital peristaltic pump. The sampler before the column helps to measure the injected tar. Samples from the trap and oil were taken for analysis at an interval of 30 minutes. Agilent 5975C VLMSD GC-MS with HP-5MS 30 m × 0.25 mm x 0.25µm capillary column analyzed the influent, effluent and oil samples. Helium was used as a carrier gas at a constant flow of 1.67x10^{-8} m^3/s. An auto-sampler injected 2x10^{-10} m^3 (0.2µl) sample to the column. Oven temperature was initially held for 4 min at 50 °C, followed by ramping to 250 °C at a rate of 5 °C/min and was finally held for 15 min at 250 ^{o}C. Oil sample was diluted 5-6 times by isopropanol. Eight k-type thermocouples (±1 °C) monitored oil temperature at the sump (T7), oil temperature at line (T0) and temperature of gas at the entrance (T1) and exit (T6) of the column and scrubbing column (4 points, T2-T5). All temperatures were logged every 10 seconds. We continuously monitored the pressure at the pump (p1) and inside the column (p2) using separate pressure gauges. A relief valve was installed before the column to reduce pressure in case of line blockage resulting in pressure build up. All the experiments were run for 4 h which was comparable to the gasifier operational time.

After completion of the experiments, we drained the oil and removed the woodchips. Around 100ml of iso-propanol poured inside the washed bottle and cleaned the column with isopropanol under the fume hood.

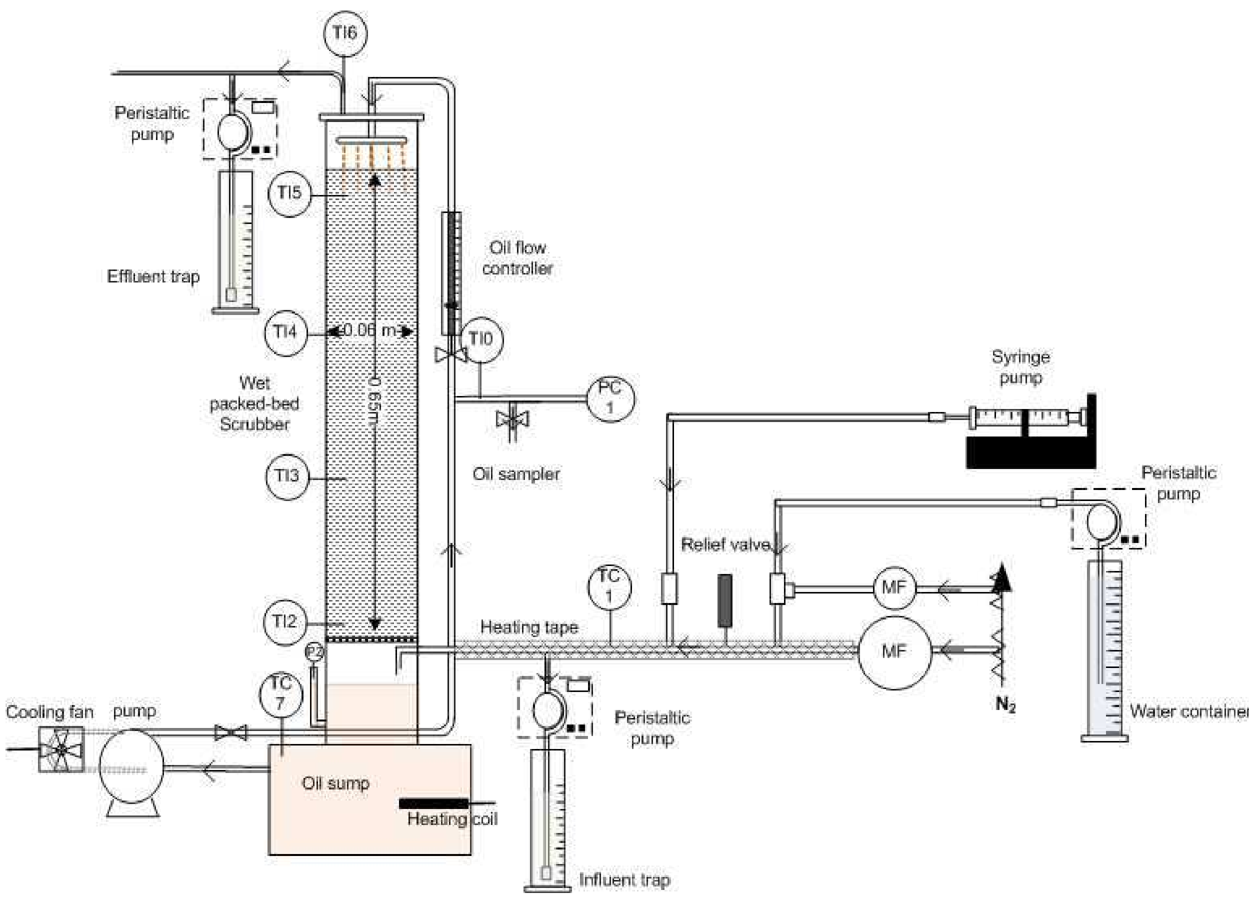

Figure 1- Schematic of wet-packed bed scrubber

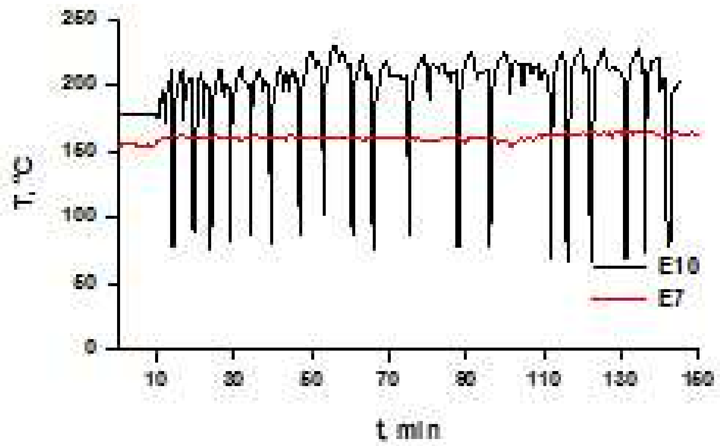

Figure 2- Temperature profile of wet packed-bed scrubber vs. time (E10: inject water in droplet form, E7: Spray water).

2.3 Design of experiments and performance efficiency

We considered liquid to gas flow ratio of ~4.7×10^{-3} and 7×10^{-3} to study the effects of oil/producer gas ratio on tar removal. Oil flow rates were $1 \times 10^{-6} m^3/s$ (60 ml/min) and 1.5×10^{-6} m^3/s (90 ml/min) while the gas flowrate was $2.17 \times 10^{-4} m^3/s$ (13 l/min). The effect of oil was studied at ambient and 50°C. All conditions were done in two bed configurations. In the first configuration the whole bed was filled with coarse woodchips (0.65m). In the second configuration 15% of coarse woodchips were replaced by fine ones. The porosity of coarse woodchips was 40% and for the fine woodchips was ~35%. Woodchips had a wide size distribution. The length, width and thickness of coarse woodchips were ~0.02-0.045 m, 0.003-0.03 m and 0.002-0.007 m, respectively. Fine woodchips had almost similar length (0.015-0.04 m) but their width (0.002-0.005 m) and thickness (0.001-0.003 m) were much smaller. We followed a statistical experimental design and named E followed by a numeral as listed in Table 2. First, we ran the experiments without loading woodchips (E9, E10). So, oil was sprayed from the top of the reactor at a flow rate of 1×10^{-6} m^3/s and contacted with the gas that flowed upward. At E9 we did not control the oil temperature. However, temperature increased from 25 °C to 35 °C gradually, because of its contact with gas at higher temperatures. In experiment E10, the oil temperature was kept at 50˚C. In the next series of experiment (E1-E8) the total length of woodchips (coarse woodchips + fine woodchips) was kept constant at 0.65 m in all experiments which was equal to 0.00184 m^3 of woodchips (Table 2). All of the experiments were done in duplicate and the mean of two values were reported.

Table 2- Design of experiments, Q_gas at 2.2 x $10^{-4} m^3/s$

No.	Q_oil x 10^6 (m^3/s)	L/G x 10^3 (m^3/s / m^3/s)	T_oil (˚C)	H_woodchips (m)	H_fine woodchips (m)
E1	1	4.7	ambient	0.65	0
E2	1	4.7	50	0.65	0
E3	1	4.7	Ambient	0.55	0.10
E4	1	4.7	50	0.55	0.10
E5	1.5	7	ambient	0.65	0
E6	1.5	7	50	0.65	0
E7	1.5	7	Ambient	0.55	0.10
E8	1.5	7	50	0.55	0.10
E9	1	4.7	Ambient	0	0
E10	1	4.7	50	0	0

To determine the efficiency of the system we used following equations:

Tar removal efficiency (RE, %) (Bhoi et al., 2015b):

$$RE, \% = \frac{T_i^{in} - T_i^{out}}{T_i^{in}} \times 100$$ Equation 1

$$RE_{overall}, \% = \frac{\sum_{i=1}^{n} T_i^{in} - T_i^{out}}{\sum_{i=1}^{n} T_i^{in}} \times 100$$ Equation 2

$i =$ Tar components; toluene, naphthalene, and anthracene

T_i^{in}= Tar component trapped at the entrance

T_i^{out}= Tar component trapped at the exit

n = number of sampling

Average tar removal efficiency

$$RE_{ave}, \% = \frac{\sum_{i=1}^{n} \frac{T_i^{in} - T_i^{out}}{T_i^{in}}}{n} \times 100 \qquad \text{Equation 3}$$

3. Results and discussions

Here we collected data, every 30 min for 4h for the results to be applicable to pilot plant gasifiers. Lab scale studies with smaller intervals for sampling (Bhoi et al., 2015b) demonstrate that the tar removal efficiency decreases considerably in the first 30-50 minute and after that the efficiency deterioration rate declines, while it stays stable at longer time (> 3h) (Nakamura et al., 2016; Tarnpradab et al., 2017) Collection of data for longer time is applicable in large scale gasifiers.

3.1 Effects of oil temperature

To study the effect of oil temperature on removing tar we inject oil at ambient and 50˚C. Gas entered into the scrubber at a temperature range of 150-160˚C and its temperature at exit was reduced to ~40˚C. In experiments E2, E4, E6, E8 and E10 the oil temperature was adjusted to 50˚C. A heater equipped with a fan kept the oil temperature at ~50˚C during the experiment. In experiments E1, E3, E5 and E7 the oil entered at ambient temperature and we did not control the oil temperature, so oil temperature increased gradually. At liquid to gas ratio of 4.7×10^{-3} the oil flow rate was lower (1×10^{-6} m^3/s), maximum temperature was close to 30˚C and at 1.5×10^{-6} m^3/s oil flow rate the oil temperature reached to 40˚C (Table 3).

Table 3- Average gas and oil temperature.

Name	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10
Tg-Influent	156.21	164.88	154.06	159.84	155.38	168.34	164.45	164.53	181.22	204.58
Tg-Effluent	42.64	44.86	44.86	45.26	36.45	44.04	36.89	44.57	45.54	48.86

T_oil-adj.	-	50.00	-	47.56	-	49.23	-	47.06	-	39.50
T_oil-ave.	31.33	49.09	31.05	48.50	38.77	48.45	39.80	47.59	34.25	42.68

In experiments E1 and E2, column was filled with coarse woodchips up to 0.65m and oil was circulated at a flow rate of 1 x 10^{-6} m^3/s. In E1, oil temperature increased gradually up to 30˚C, while in E2 we controlled the temperature at 50 ˚C. Comparing E1 and E2, Increasing the temperature from ambient to 50˚C reduced toluene and naphthalene removal efficiencies 3% and 7%, respectively. E5 and E6 had the same bed configuration as E1 and E2, but with the oil flow rate increased to 1.5x10^{-6} m^3/s- (Oil/gas ratio =7 x 10^{-3}). At E5 and E6, increasing the temperature from ~23˚C to 50˚C reduced average toluene removal efficiency from 62% to 56%. Toluene and naphthalene removal efficiency diminish 5% and 13% by increasing the temperature from ambient to 50˚C (Figure 3). Temperature has higher effect on naphthalene removal rather than toluene. Because the height of the bed is low (65cm), the effect of temperature is not dominant. The effect of temperature is more pronounced at higher bed height and higher flow rate. For example, the effect of vegetable oil temperature was not prominent at 0.5m bed height; while, it was significant at higher heights (0.8m and 1.1m) (Bhoi et al., 2015b).

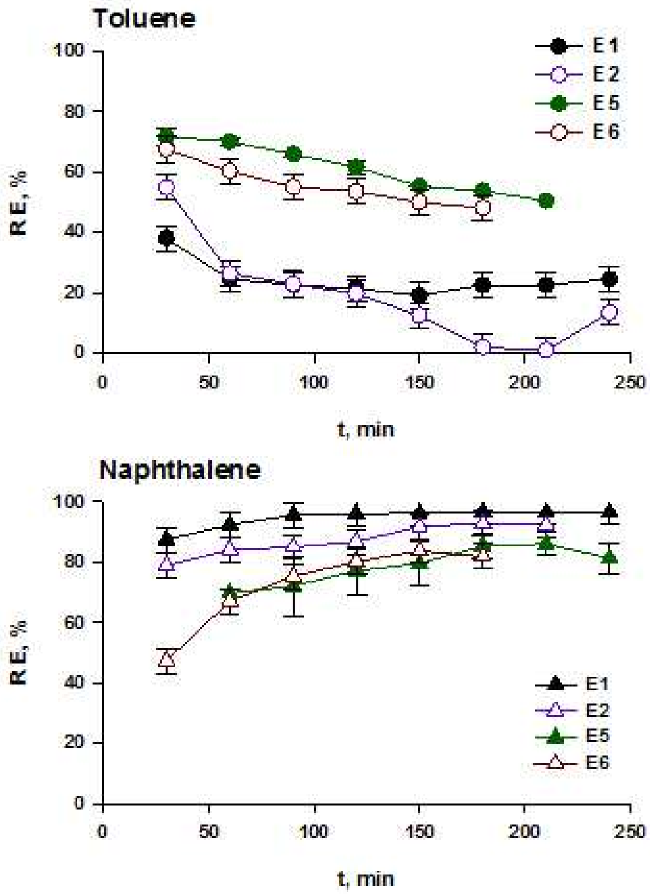

Figure 3- Effect of oil flow rate and temperature on the removal efficiency of toluene & naphthalene (oil/gas_E1&E2= 4.6x10^{-3}; oil/gas_E5&E6=7x10^{-3}).

Temperature of solvent affects the tar removal efficiency significantly by affecting the viscosity of solvent and consequently mass transfer coefficient. Oil temperature can have both positive and negative effect on tar removal efficiency. Our results demonstrate its negative effect is dominant compared to the positive effect. Tar compounds are more soluble at lower temperature due to lower density, viscosity and surface tension (Bhoi et al., 2015b). Gas temperature affects tar condensation which plays a key role in removing tar (Thapa et al., 2017). As tar condensation has a considerable role in removing naphthalene (Paethanom et al., 2013), by Increasing the temperature less naphthalene condensed on the wood surface. Increasing the temperature reduces the viscosity and improves oil spraying quality, as well. Miniaturization of bio-oil improves contact area between gas and liquid and accordingly the mass transfer. While, cooling the gas has negative effects on tar removal (Nakamura et al., 2016). Woodchips imbibe more oil, so the effect of temperature is more observable when metals or plastics are used as a packing material compared to woodchips. Absorption of the oil causes woodchips to inflate, so their surface area increases. Higher surface area causes more naphthalene condensation and removal (Figure 3). It could be the reason that RE_naphthalene increased. For toluene, tar removal efficiency increase to some extent after 160min (Figure 5). In this system, adsorption, absorption and condensation affect tar removal efficiency.

3.2 Effects of oil/gas ratio

Oil to gas ratio had a significant effect on tar removal efficiency of the toluene as a tar model compound (Figure 4). By increasing the oil flow rate from 1×10^{-6} to 1.5×10^{-6} m^3/s at T_oil,ambient RE_toluene increased from 20% (E1) to 62% (E5) (a removal efficiency difference (Δ) of 42%) and at T_oil of 50˚C, RE_toluene from 18.9% (E2) reached to 55% (E6) (Δ=36%). Increasing L/G from 4.7×10^{-3} ($Q_{oil}=1 \times 10^{-6}$ m^3/s) to 7×10^{-3}($Q_{oil}=1.5 \times 10^{-6}$ m^3/s) had an opposite effect on RE_naphthalene, but, it was not as significant as its effect on toluene (Figure 3). RE_{ave} naphthalene reduced from 94% to 72% (Δ=-22%) and from 87% to 72% (Δ= -15%) when oil to gas ratio increased from 4.6×10^{-3} to 7×10^{-3} times at ambient and 50˚C, respectively (Figure 5).

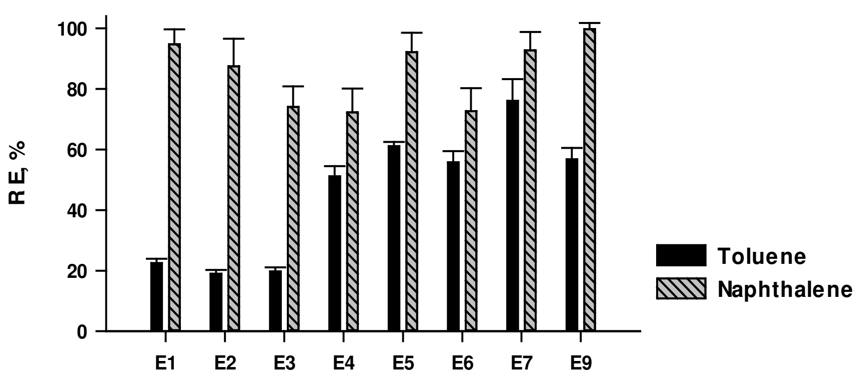

Figure 4- Average removal efficiency (L/G_ E1-4, 9= 4.7×10^{-3}, L/G_E5-E7= 7×10^{-3}).

In experiment E3 and E7 where 0.1 m of woodchips was replaced by fine woodchips increasing oil/gas ratio from 4.6×10^{-6} to 7×10^{-6} improved RE_toluene ~65%, from 19% to 86% [3]. In these experiments, increasing oil flowrate enhanced RE_naphthalene ~16%, which is opposite of the previous observations (comparisons of E1&E5 and E2&E6) (Figure 5). After the experiment and opening the column, we observed part of the coarse woodchips remained dry at E3. So, the presence of oil has a positive effect on naphthalene removal efficiency, but L/G should be optimized. Increasing the liquid flow rate will increase the driving force for the mass transfer, but simultaneously will reduce the cross sectional area of the column and consequently increase the gas velocity. Also, it increases liquid film thickness that leads to higher mass transfer resistance to liquid side. So, these parameters should be optimized. Oil residue analysis demonstrates most of the removed tar is trapped in oil. The rest is adsorbed or condensed on woodchips (Table 4).

Table 4- Concentrations of toluene and naphthalene in oil residue.

Name	**toluene, %**	**naphthalene,%**
E5	49.12	80.25
E6	44.01	NA
E7	61.19	90

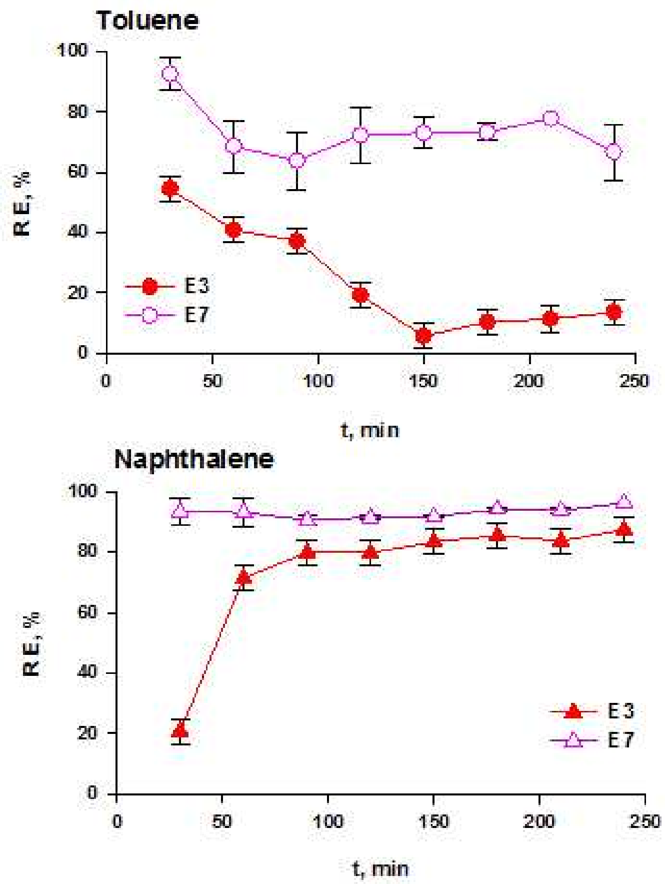

Figure 5- Effect of oil flow rate on tar removal efficiency (L/G_E3 = $4.6x10^{-3}$; L/G_E7 =$7x10^{-3}$).

Table 5- Adsorbed oil in woodchips.

Experiments	Adsorbed oil, wt%	
	woodchips	Fine woodchips
E1	_	
E2	10.9	
E3	_	_
E4	_	_
E5	31.9	60.4
E6	41.25	
E7	43.8	38.6
E8	40.6	49.2

Part of the oil adsorbs in woodchips and increase their weight by around 40% (Table 5). Weight of oil increased 8%, 5.7%, 5.8% and 2% at E5, E6, E7 and E8, respectively which demonstrates that higher flow rate not only improves tar removal efficiency, but also causes more water traps. Increasing the oil temperature reduced the water content to some extent.

3.3 Effects of bed size distribution

Comparing experiments E5 and E7 ($Q\_oil = 1.5x10^{-6}m^3/s$ and $T\_oil\_in = 25^\circ C$), replacing ~15% of woodchips (0.1m) with fine ones increased RE_{ave} of toluene and naphthalene 24% and 18%, respectively. Replacement of coarse woodchips with fine ones improves, both, absorption that is good for light tar compounds and adsorption that is suitable for heavy tar compounds. Fine woodchips increase the contact time of toluene with oil and cause RE_{ave} toluene increase. Higher surface area of fine woodchips improves condensation of heavier tars like naphthalene. In experiment E3, we kept all the conditions similar to E1 ($Q\_oil = 1x10^{-6}m^3/s$ and $T\_oil\_in = 25^\circ C$), but replaced 15% of coarse woodchips with fine ones. We observed RE_{ave} of toluene and naphthalene reduced 2.7% and 20.7% which was opposite of our observation in E5 and E7. Experiments E2 and E4 had the same Q_oil ($1x10^{-6}m^3/s$) and $T\_oil = 50^\circ C$, but in E4, ~10cm of coarse woodchips were replaced with fine ones. At E4, similar to E3, RE_{ave} of naphthalene reduced from 82% to 72% (Δ=15%), but, RE_{ave} of toluene increased. After the experiments and disassembling the setup, we observed more than 70% and 90% of the coarse woodchips remained dry in E3 and E4, respectively. Fine woodchips were completely wet but as most part of the bed remained dry; so, gas was not in contact with oil through part of the bed. It demonstrates oil has considerable effect on tar removal efficiency rather than woodchips. Comparison of E3 and E4 with E9 in Figure 4 also confirms the importance of oil on removing tar. In E9 only oil contacted with gas, toluene removal efficiency is 56%, while RE_{ave} of E3 and E4 are 20% and 51%, respectively. Thapa et al. (2017) also demonstrated bubbling the producer gas inside oil before passing it through a wood shaving filter increased tar reduction efficiency from 10% to 97% (Thapa et al., 2017). Oil absorbs the heavy tar compounds. The light tars that remained in gas condense at cold woodchips, wood shavings or rice husk char as a filter (Paethanom et al., 2012).

At higher oil flow rate, the toluene and naphthalene removal efficiencies were in the range of 60-86% and 79-90% in our wet packed-bed scrubber, which is comparable to the previously reported results.

3.4 Statistical analysis

We studied the effect of temperature, liquid flow rate (L/G ratio) and bed distribution as the main three parameters on tar removal efficiency. To demonstrate their influence, we calculated p-value using ANOVA at Minitab software. At ANOVA, we used a general linear model that gives us more flexibility, and we considered RE as the response and the studied parameters as factors. P-value measured the strength of the evidence against the null hypothesis and its approach was to reject the null hypothesis. So, smaller amount of p-value means that the parameter has a more significant effect on tar removal efficiency.

Table 6- P-value-Effect of temperature, L/G ratio and bed distribution on tar removal.

p-value	Temperature	oil flowrate	bed configuration
Toluene	0.24	0.002	0.014
Naphthalene	0.34	0.022	0.024

Based on the p-value, oil flow rate has the main impact on tar removal, especially toluene removal. In removing the toluene, L/G ratio has significant effect (p-value= 0.002). After that, bed configuration affects the toluene removal (p-value= 0.014). For naphthalene, both, oil flow rate and bed configuration have almost similar effect with p-value of 0.022 and 0.024 respectively. The effect of oil temperature was lower compare to oil flowrate and bed configuration (Table 6). Temperature reduces density and wettability of oil and consequently its capability in removing tar, so, the effect of temperature is more observable when metals or plastics are used as a packing material compare to woodchips. At lower packing-bed height, the effect of temperature is lower.

4. Conclusion

A wet packed-bed scrubber was employed to investigate the efficiency of woodchips and waste cooking oil on removing tar. L/G ratio had the most considerable impact on tar removal efficiency. Packing bed size distribution influence on tar removal efficiency (RE) was not significant. The effect of oil temperature on tar RE was less than L/G ratio. The highest RE among the studied conditions was obtained at L/G ratio of 7×10^{-3}, when oil temperature was 25 °C and 15% of woodchips were replaced with fine woodchips. The system was able to remove 70% and 90% of toluene and naphthalene, respectively.

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