

Review Paper

Tar removal from Producer Gas: A Review

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Abstract

Gasification is the most appropriate technology for conversion of solid fuel (biomass) into a gaseous fuel, known as producer gas. Producer gas is a mixture of gases which consists of hydrogen, carbon monoxide, methane, carbon dioxide, water vapor, nitrogen, tar and suspended particulate matter. For motive applications such as internal combustion engines, the tar present in producer gas may create problem, if the tar content in the producer gas is above 50-100 mg/Nm³. A tar-free gaseous fuel can be obtained in a suitably designed producer gas conditioning unit whose sole purpose is to provide clean producer gas. Gas cleaning and conditioning systems to control tar levels are being continuously modified for better efficiency and cost effectiveness. Major techniques used in tar cleaning are thermal cracking, catalytic cracking and physical removal of tar. Many a times, combination of these techniques are used for better cleaning of producer gas. The following paper critically reviews the different techniques used for collection, identification and quantification of tars in producer gas obtained from biomass.

Keywords: Biomass, Tar, producer gas cleaning, catalytic cracking, gasification, suspended particulate matter.

Introduction

Due to environmental concern there is an increasing world-wide interest in the use of biomass resources as a feedstock for fuels and chemicals. Biomass resources are one of the most important components to eliminate global climatic change. Plant growth; recycle CO₂ from the recycle atmosphere and the use of biomass resources for energy and chemicals results in low net emission of carbon dioxide. This helps biomass technology to meet local and regional environmental regulations and reduce emissions that contribute to acid rain. The use of these locally produces energy resources which results in new market for agriculture and forestry products and provides a mechanism for rural economic development. Because of these and other factors, many governments are currently developing policies and regulations intended to expand the use of biomass over the next decade and beyond. Biomass gasification technologies are

estimated to be an important part of the effort to meet these goals of intensifying the use of biomass. Gasification technologies provide the opportunity to convert renewable biomass feedstock into clean fuel gases or synthesis gases. These gaseous products can be burned to generate heat or electricity, or they can potentially be used in the synthesis of liquid transportation fuels. Gasification offers a combination of flexibility, efficiency and environmental acceptability that is essential in meeting future energy requirements. In a biomass gasifier, the carbonaceous fuels are reacted with air/oxygen resulting in combustible gas. When a pyrolysis fuel (wood) is used, hydrocarbon (tar) will be evolved which have a condensation temperature of less than 150°C. Tars are condensable portion of the organic gasification products and are largely aromatic hydrocarbons, including benzene¹ (figure-1).

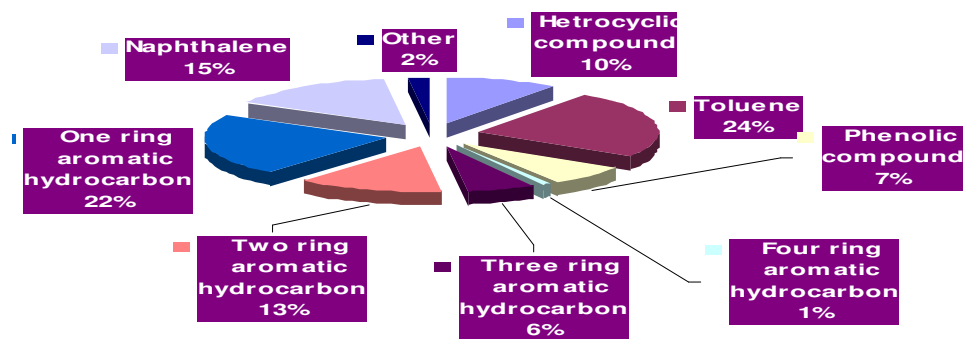


Figure-1
Typical composition of biomass tars²⁹

Tar sampling protocols are being developed by Simell et al and Neeft et al^{2,3} to help in standardize the way tars are collected; however, these methods are not yet widely established. Regardless of how “tar” is defined, tar removal, conversion or destruction is seen as one of the greatest technical challenges to overcome the successful development of commercial advanced gasification technologies. The allowable tar levels are about 50, 5, 1 mg/Nm³ for gas engines, gas turbines and fuel cells respectively⁴.

As reported by Milne et al each type of gasifier has different reaction conditions and consequently different tar compositions and production rates. Reported tar yields range from 10 to 100 g/Nm³ of gas for updraft gasifier and 50–500 mg/Nm³ of gas for downdraft gasifier. It was also observed that tar content in producer gas varied from 0.04 to 150 g/Nm³ depending upon gasification reactor (Table-1)⁵.

Table-1
Comparison of measured tar levels from different biomass gasifier design²

Gasifier Type	Tar Loading g/Nm ³		
	Min	Max	Representative Range
Fixed Bed			
Downdraft	0.04	6	0.1 - 1.2
Up-draft	1	150	20 - 100
Moving Bed			
Fluidized Bed	< 0.1	23	0.9 - 15
Circulating Fluidized Bed	< 1	30	0.9 - 15

Required gas cleanliness standards for successful gasifier-engine systems are recommended from 10 mg/Nm³ to less than 1mg/Nm³ (table-2). Thus, even the best design of conventional downdraft gasifier produces tar that is 50 times higher in concentration than is desirable. Even after elaborate cleaning producer gas normally has tar content well above the desirable upper limit. Tar concentrations in cleaned gas using highly efficient scrubber has been reported to be as 70–75 mg/Nm³.

Table-2
End use device tar tolerances¹²

End Use Device	Tar Tolerances (mg/Nm ³)
SI and Diesel Engine	10 - 100
Industrial Gas Turbines	< 5
Compressors	50 - 500
Fuel Cells	< 1

The aim of this paper is to study the tar removal technologies, methodology and problems associated with biomass devices. An actual evaluation of the data available for tar in the literature, is not easy because of the use of dissimilar operating conditions, different type of gasifier used, variable tar capturing, sampling

and analyzing method, and most prominently the non-consistency in defining tar used by various researchers.

Tar Removal Approaches: Generally two approaches: Primary method and Secondary method was used for Tar removal.

Primary Treatment Approaches: It is a method which reduces the tar content in the producer gas and is employed inside the gasifier without the need of a secondary reactor, it is done by: i. Proper selection of operating parameters (temperature, gasifying agent, equivalence ratio, residence time). ii. Use of bed additives/ catalyst. iii. Gasifier Modification.

Proper selection of operating parameters: Zhao et al studied the effect of fuel feeding on tar. In the reactor fuel was fed in two stages: one from top and another from lower part of the gasifier. It was observed that tar reduction was more (2.39 g/Nm³) in two stage feeding compared to single stage feeding (2.15 g/Nm³). It may be due to two stage feeding, endothermic heat helps in fuel pyrolysis which reduces the tar content⁶. Mayerhofer et al studied the effect of oxidizing agent/waste ratio, and found that the tar yield could be decreases by increasing this ratio⁷. Yi Su et al observed that on a continuous reactor system at partial oxidation environment, oxygen has a huge impact on the conversion of tar and condensable gases. The amount of tar reduces rapidly with equivalence ratio and when equivalence ratio was 0.34, the tar reaches the least values of 0.26%⁸. Kitapong et al⁹ compares the tar content at throatless downdraft gasifier with singles and double air supply position and found that the tar content decreases from 114 to 43.2 mg/Nm³. Yu-Hong Qin et al while working on saw-dust in a fluidized bed reactor under air-steam gasification found that adding up steam and increase of temperature there will be a decline in tar yield. This is due to the influence of steam on high molecular mass component which increases at elevated temperature. They also found that steam mainly influence the composition of molecular mass from 130 to 200 AMU (atomic mass unit) compounds and makes the tar components aromaticity decrease at 800°C. The steam does not alter the molecular mass distribution, but the relative amount of each fraction does change and if the part of steam to biomass increased appropriately the condensation of tar can be avoided¹⁰. Phuphuakrat et al studied the effect of ER on tar using dried sewage sludge in a downdraft gasifier, and found that at higher ER significant amount of tar could be reduced¹¹. Han and Kim found that higher temperature and long residence time, helped in cracking of tar into lighter molecules¹². Montri et al found that the tar concentration will reduce with increasing temperature, decreasing bed particle size, and increasing residence time under controlled supply of air¹³. Wang et al observed a decrease in the amount of light hydrocarbon as well as that of tar in the fuel gas with an increasing equivalence ratio for pressurized gasification with 100% carbon conversion¹⁴. Passen and Hiel studied the impact of feed stock properties and gasifier operating condition on tar development. The gasification temperature was increased from 750°C to 900°C with steps of

50°C, and maintaining the constant equivalence ratio. They found that Solid Phase Absorption (mg/m^3) increased from 725°C to 760°C temperature latter it remains constant up to 800°C and then reduce gradually up to 900°C. The class 2 tars are almost decomposed at a gasification temperature of 850°C, however the class 4 and heavy class 5 tar gradually decomposed maximum at 780 – 800°C. This shows that the gasification temperature appears to have huge impact on the tar composition¹⁵. Li X et al, reported that tar yield from biomass gasification decrease drastically from 15 g/Nm^3 to 0.54 g/Nm^3 as the average temperature amplify from 970 to 1070°C¹⁶. Lapamudra Devi et al reported, more than 40 % reduction in tar yield could be achieved when the temperature is raised from 700°C to 900°C. They also reported that residence time has slight influence on the tar yield, however it appreciably influence the tar composition¹⁷. Fagbemi et al found that tar yield increase with the increase in gasification temperature up to 600°C and then start reducing with further temperature increment. This phenomenon can be explained as, when the temperature is higher than 600°C the secondary reaction prevails which leads to tar breakdown. Equivalence ratio increment also has a positive effect on tar formation, but the heat value will decrease with equivalence ratio¹⁸. Bhattacharya et al worked on two-stage gasification with preheated air and found that tar in producer gas was significantly low with preheated air supply and was below 10 mg/Nm^3 ¹⁹. Pan et al while working with fluidized bed gasification observed that injecting secondary air at temperature over 830°C – 850°C reduces the tar content. It shows that the formation of tars significantly depends on operating condition. When the temperature increases from 800°C to about 830°C tar content of the forest waste decreases abruptly from 6.7 g/Nm^3 to 4.7 g/Nm^3 and further increase in temperature up to 1000°C, reduces the tar content to 2.5 g/Nm^3 ²⁰. Narvaez et al studied biomass gasification at different temperatures and found that the tar content at 700°C and 800°C were 19 g/Nm^3 and 5 g/Nm^3 respectively^{21, 22}.

Kinoshita et al reported that tar yield and its concentration decrease as equivalence ratio is increased. This is because of more availability of oxygen to react with volatiles in the flaming pyrolysis zone. They also found that residence time has little influence on the tar yield, however it significantly influence the tar composition²³.

Critical analysis of operating parameters such as temperature, gasifying agent, equivalence ratio, and residence time est. reveals that although number of the parameters are responsible for efficient removal of tar from producer gas, however reactor temperature has a major role.

Use of bed additives / catalyst: Toshiaki et al studied the effect of iron and activated carbon for removal of tar from producer gas and found that tar could be reduced to 1/24 of its concentration i.e. from (2428 mg/Nm^3 to 102 mg/Nm^3)²⁴. Jin-Won Kim et al used activated carbon and dolomite for tar removal and observed that activated carbon was better than

dolomite for tar removal. The amount of additive was also a vital factor for efficient tar deduction. It was also noted that 640g of activated carbon in the upper-reactor could drastically reduce the tar (50.99 g/Nm^3 – 19.25 g/Nm^3)²⁵. At standard temperature and pressure magnesite as bed material could reduce the tar (@ 2 g/Nm^3)²⁶. Similarly under a controlled temperature and air flow rate palladium (Pd) as a hydro-cracking catalyst could reduce tarry component up to 99% in a updraft gasifier²⁷. Rapagna et al developed a catalyst with a chemical formula of $\text{LaNi}_{0.3}\text{Fe}_{0.7}\text{O}_3$ that was prepared by means of a sol-gel related process where La, Ni, and Fe nitrate salts were dissolved separately in hot propionic acid. This catalyst was tested as a secondary catalytic reactor operating at 800°C downstream of a fluidized bed gasifier. Almond shells were gasified at 770°C in a bed of olivine with a steam: biomass ratio of 1:1, a 90% tar reduction was measured²⁸. Coll R et al compared five catalysts (benzene, toluene, naphthalene, anthracene and pyrene) on tar removal from fuel gases in a fixed-bed reactor. Y-zeolite and Ni-Mo catalyst were found to be most effective, as it can remove almost 100 % tar at 550°C temperature. They also observed that process variables like temperature and space velocity had very major effect on tar removal²⁹. Corella et al reported that the use of calcined dolomite on the inner side of the gasifier reactor could reduce the tar quantity from 6.6 wt% (without dolomite) to 1.3 wt%. (With dolomite)³⁰. Studies conducted by Rapagna et al reveals that the presence of dolomite in the fluidized bed had the advantage of reducing tar content, however dolomite could not affect gaseous hydro-carbon concentration. It was reported that an amount of 20-30 wt% dolomite crest being silica sand in the gasifier reduce tar content @ about 1 g/Nm^3 at an equivalence ratio of 0.3³¹. Chembukulam et al suggested that cracking over a char bed at a temperature of 950°C resulted in nearly complete decomposition of tar³². Studied conducted by various researcher on the effect of catalysts like dolomite, limestone, olivine sand, bauxite, lanthanum, alumina, nickel aluminate, cobalt, natural clay minerals and iron minerals on tar reforming at high temperature, suggest that although it is an efficient method for the tar destruction, however this primary method may be very expensive in function^{33, 34, 35, 36, 37, 38}. Felice et al (2010) studied the effect of iron as a catalytic activity and found that both Fe^{3+} and Fe^{2+} in assistance with CaO and MgO substrates are active in tar-reforming reactions⁴¹.

Many catalyst and additives were investigated by different researchers for the removal of tar but activated carbon, calcined dolomite and Y-zeolite at 550 °C is best suited for tar removal.

(c) Gasifier Modification: Pan et al reported that introduction of secondary air just above the biomass feeding point by 20% to primary air at 840°C – 880°C will reduce the total tar concentration by 88.7 wt %⁴⁰. The studied carried by Asian Institute of Technology (AIT) Thailand (1998) on two stage gasification suggested that clean gas producer gas could be obtained by separating of pyrolysis zone and reduction zone of

gasifier, as tars formed during the pyrolysis zone could be decomposed in the reduction zone.

Narvaez et al reported that with longer size of gasification, the temperature rise in the freeboard of fluidized-bed gasifier due to air injection could be higher and a temperature rise of about 70°C would reduce the tar from (28 g/Nm³ to 16 g/Nm³)^{21, 22}.

Provision of secondary air in the reactor would increase the temperature of the gas which will reduce the tar concentration.

Secondary Treatment Approaches: It is a method which uses a separate reactor to reduce the tar content in producer gas up-to acceptable level. Secondary tar cleaning techniques could be further divided as wet and hot gas cleaning.

Wet Cleaning: Wet gas cleaning uses water scrubbing and venture scrubbing to condense the tar compounds from the producer gas and simultaneously removing the particulates. Adi and Fajri studied the effect of venture scrubber on producer gas tar obtained from downdraft gasifier and found that by increasing the water flow rate of venture, tar content of the producer gas could be reduced. Bergman et al developed a wet scrubbing (scrubbing liquid was other than water which was regenerated and recycled) based cleaning technology called “OLGA”. They reported that by removing tar components (heavy poly-aromatic hydrocarbons (≥4 rings), light poly aromatic hydrocarbon (2–3 rings) and heterocyclic compounds) cleaned producer gas, acceptable for most motive power application could be obtained. These Tar components, condensed above 25 °C, based on their dew points⁴². Baker et al reported that producer gas with tar concentrations below 20 – 40 mg/Nm³ can be achieved using a venture scrubbing system (table-3)

Table-3
Tar removal efficiencies of wet scrubbers²

Technology	Tar Removal Efficiency
Spray Tower	11 - 25 % Heavy Tars
	40 - 60% PAH
	0 - 60% Phenols
Venturi Scrubber	50 - 90 %
Venturi and Spray Scrubber	83 - 99 % Condensable material
Venturi + Cyclonic Demister	93 - 99 % Condensable organics
Vortex Scrubber	66 - 78 % Evaporation residues

This technology has been used extensively in coke-oven and gas processing industries (figure-2).

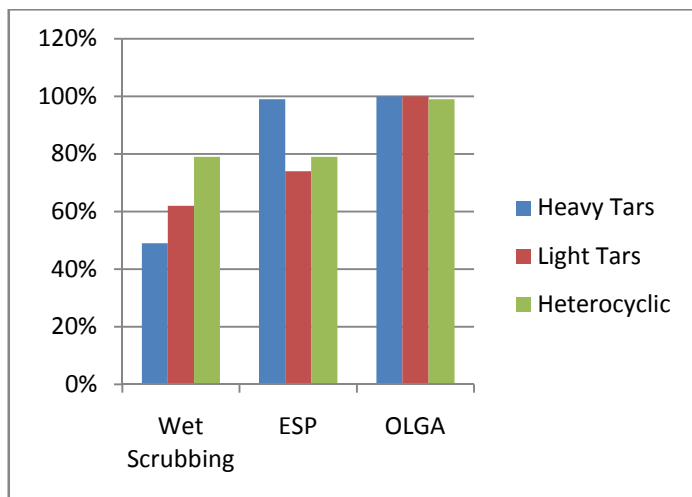


Figure-2
Removal Efficiency of different technologies⁴²

The exit gas temperature from wet cleaning methods was 35–60°C, which results in loss of sensible heat and water condensate requires treatment before disposal. The use of barrier filters and cyclone separators have not been effective for tar removal as tar aerosol particles are less than 1µm in size and are sticky in nature which makes them difficult to remove from walls of the cyclone and filter. Electrostatic precipitator (ESP), wet scrubbers can remove most of the tar up to about 150 °C, however more expensive. The tar collected using these physical techniques can be burned to produce heat or re-injected into a gasifier⁴³.

Wet cleaning of producer gas is one of the effective methods for removal of Tar and particulate from producer gas, however it also creates environmental problems by leaving wet effluent.

(b) Hot Gas Cleaning: The objective of hot gas cleaning is to crack the tar by applying high temperature. Teeranai et al studied the effect of intra-particle on wood and sawdust during the process of pyrolysis and found that below 380° C both the woody biomass release the same amount of volatile matter but at 400 – 500° C decomposition of intraparticle to secondary air will occur which will acts as a catalyst and will help in reducing the tar content of producer gas generated from large woody particle⁴⁴. Jordan and Akay studied the composition of tar generated during gasification by using fuel cell bagasse (FCB) in a downdraft throat type gasifier to evaluate their dew point temperature and found that class 2 (heterocyclic aromatic) and class 5 (heavy polyaromatic) which dominates the tar composition will condense at low concentration of (0.1 mg/m³)⁴⁵. Young et al studied on the tar removal procedure through 3 phase arc plasma and found that it has a high breakdown efficiency of tar along with effortless reaction controlling and high energy effectiveness⁴⁶. Non-catalytic reforming performed by Wang et al at 800–950°C indicated that supplying the air to the reformer decrease the high heating value of the producer gas by partial combustion and dilute the

reformed gas⁴⁷. Huber et al investigated that without a catalyst, the temperature required for tar cracking is above 850°C which reduces efficiency, causes material problems and produces soot⁴⁸. Inaba et al compared two types of supports for production of hydrogen over Ni catalysts and found that metal oxides produced large quantities of dark-colored tar while zeolite-based support produced carbon deposition without tar formation. Higher temperatures led to higher rates of gasification and H₂ production, and decreased depositions of tar and char on the catalysts⁴⁹. Han and Kim stated that hot gas cleaning results in increased hydrogen production in the producer gas because the destruction of tar yields hydrogen, at the higher temperature and catalysts provide favorable conditions for reforming and shift reactions¹². Aznar and Corella reported that catalysts from naphtha were more effective in destructing tar than catalysts from natural gas reforming. Operating conditions such as temperature, space time (mass of catalyst per unit flow rate of producer gas), catalyst particle size and gas compositions affected tar conversion efficiencies, they suggested that increase in equivalence ratio from 0.15 to 0.21, tar concentration reduces from 5.7 to 1.0 g/Nm³ and by supplying steam (up to Steam/Biomass of 0.5) to the reformer, increased the high heating value and cold gas efficiency of the reformed gas. H₂ and CH₄ contents increased but CO and tar contents decreased with increasing steam/biomass into the reformer. Supplying steam or/and air to the secondary reactor, in presence of catalysts, enables the tar to react and form CO, H₂, CO₂ and CH₄. They also reported that activity of Ni/dolomite catalysts in the secondary catalytic reactor were comparable to commercial steam reforming catalysts. Ni / dolomite catalysts are comparatively cheaper than steam reforming catalysts and have anticoking properties. With a space time of 0.02 kg of catalyst (m³/h) and a temperature of 850°C, the catalysts were able to convert 98% of the tar while increasing H₂ content by comparing commercial steam reforming catalysts for naphtha and natural gas⁵⁰.

It is observed that hot gas cleaning (500°C to 900°C temperature) may reduce the tar content up to the strength of 98% and also increase the calorific value of gas by destruction of tar in to hydrogen. However due to need of high temperature, complete package becomes uneconomical.

Conclusion

Critical review of literature reveals that number of technology/process has been developed for tar removal from producer gas, however no technology/process enjoy the acceptance of universe due to variation in characteristics of biomass as well as design of gas producer. Physical separation process will continue to play a very important role for the successful commercial implementation of gasification. Tar present in producer gas is removed mainly through wet or dry scrubbing, as it could be easily designed and applied depending on the specific need of any gasification process. Although there

is need to design of highly efficient wet or dry scrubber to minimize the waste water or solid residues.

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