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Technologies for Tar Removal from Biomass-Derived Syngas

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Abstract

The concerns on the significant negative environmental impacts of conventional fuels such as coal, oil and natural gas have fostered the shift of energy consumption towards renewable and environment friendly sources like biomass derived energy. Many technologies have been developed to generate energy from biomass; among them gasification is considered one of the most promising technologies, since and the generated syngas has many practical applications, such as a world-wide sustainable energy production or synthesis of fuels and chemicals. Unavoidable produced impurities during gasification can create severe problems in downstream applications; therefore, the cleaning of the produced syngas is essential. A major challenge in commercialization of syngas technology and its valorization is tar removal method. This review organizes the knowledge related to tar generated from biomass-derived syngas, and discusses the recent progress on clean technologies for tar elimination. The advantages and disadvantages of different tar removal methods are critically discussed. Primary treatment is able to optimise the gas composition for the secondary cleaning step but is not sufficient for further syngas applications. Among the secondary treatments, the catalytic cracking is most valued in terms of energy and yield efficiencies for syngas cleaning. This review discusses current technical barriers and future opportunities of technical development.

Keywords: Biomass; Gasification; Tar removal; Syngas; Primary methods; Catalytic conversion; Economic view

Abbreviations: AIT: Asian Institute of Technology; BFBG: Bubbling Fluidized Bed Gasifiers; BTX: Benzene/Toluene/Xylene; CHP: Combined Heat and Power; CHRIGAS: Clean Hydrogen-rich Synthesis Gas; ECN: Energy Research Center of the Netherland; ER: Equivalence Ratio; ESP: Electrostatic Precipitators; GR: Gasifying Ratio; IG: Integrated Gasification; LHC: Light Hydrocarbons; PAH: Poly-Aromatic Hydrocarbons ; RF: Radio Frequency; RSP: Rotating Particle Separators; SB: Steam–Biomass Ratio

Introduction

Global warming and climate change have encouraged researchers to look for an alternative to replace fossil fuels. Replacing biomass with fossil fuels reduces greenhouse gas environmental impact and offers a solution to waste [1]. Energy from biomass is created either by direct burning or thermochemical or biochemical conversion into liquid or gaseous fuels. An advantage of thermochemical processing instead of biochemical processing and direct burning is that it can more readily break down biomass in a controlled manner to produce high concentrations of desired intermediates [2]. The two primary thermochemical approaches to convert biomass into fuels are gasification and pyrolysis. Biomass gasification technology that converts biomass to synthesis gas (syngas) has been investigated thoroughly to convert the low value and highly distributed solid biomass to a uniform gaseous mixture mainly including hydrogen (H_2), carbon monoxide (CO), methane (CH_4), and carbon dioxide (CO_2) [3]. The application of syngas includes power generation using internal combustion (IC) engines, gas turbines, especially in remote areas with no electricity supply, fuel cells, and the synthesis production of liquid fuels and chemicals. Although many advantages of biomass gasification and wide application of syngas, commercial acceptability of technology still faces challenges due to the difficulty in cleaning the unwanted by-products (Figure 1), such as particulate matter, condensable hydrocarbons (i.e., tars), sulfur compounds (H_2S and SO_2), nitrogen compounds (NH_3), alkali metals (primarily potassium and sodium), and hydrogen chloride (HCl) (Table 1) to meet process requirements and pollution control regulations (Table 2) [4,5].

$$
\begin{aligned}
&Biomass + O_2\left(or\ H_2O\right) \rightarrow CO, CO_2, H_2O, H_2, CH_4 + other\ hydrocarbons\\
&\rightarrow tar + char + ash\left(organic\ contaminants\right)\\
&\rightarrow HCN, NH_2, HCl, H_2S + other\ sulfur\ gases\left(inorganic\ contaminants\right)\\
&\rightarrow particulates
\end{aligned}
$$

Figure 1: Scheme of biomass gasification [6].

Among them, tars that constitute 0.1–10% of produced gas recognized as a universal challenge of gasification system and gain priorities for removal, because of condensation and subsequent plugging — it can potentially foul filters, lines, and engines, as well as deactivate catalysts in cleanup systems or downstream processes, resulting in serious operational interruptions [4,7-9]. They also cause ineffective energy conversion since tar can polymerize into more complex structures which contain significant amounts of unused chemicals that could have otherwise been transformed into useful fuel gases such as H_2, CO, CH_4, etc. Additionally, formation of tar aerosol is toxic since it contains compounds with carcinogenic character. Up to now, a great amount of work on tar removal or reduction and comprehensive reviews have been reported. The present review updates and complements those past reviews by extending the review to discuss tar removal technologies at demonstration and pilot scale.

Contaminant	Presence	Problems
Particulates	Derive from ash, char, condensing compounds, and bed material of fluidized bed reactors	Metallic components erosion, and environmental pollution
Alkali metals	Presence of alkali metals compounds, specially sodium and potassium in vapor phase	Metal corrosion at high-temperature due to the stripping off of the protective oxide layer
Fuel-bound nitrogen	Forming NO_x during combustion	NO_x emission
Sulfur and chlorine	Not considered to be a problem in the biomass and waste	Harmful pollutants, and acid corrosion of metals
Tar	It is bituminous oil constituted by a complex mixture of oxygenated hydrocarbons existing in vapor phase in the producer gas, it is difficult to remove by simple condensation	Filters and valves clogging, and metallic corrosion

Table 1: Contaminant presence in the gas and relative problems [4].

Application	Heating value (MJ-Nm^3)	Syngas composition(mg-Nm^{-3})			
		Dust	Tar	H_2S	Alkalis
Combustion	>4	no limit			
Engine		5–50	<100	<700	1–2
Gas Turbine		<5	<5	<1	<0.2
Fuel Cell		<0.1	<1	<0.1	<0.1
Methanol production		<0.1	<1	<0.1	<0.25

Table 2: Gas quality requirements for different applications [10,11].

Tar Definition and Classification

Biomass in the presence of oxygen or steam is gasified and produces syngas, organic and inorganic contaminants and particulates (Figure 1). Tar forms during the condensation of syngas, which is a progress from highly oxygenated compounds of moderate molecular weight to heavy, highly reduced compounds. Hundreds or even thousands of different tar species are generated during the gasification and condensation, because of different operating parameters, thermochemical conversion processes. Feedstock composition, processing conditions, especially temperature (Figure 2), pressure, type and amount of oxidant, and feedstock residence time strongly affect the tar composition [12], which results in difficulties in collecting, analyzing, and even defining them. A recent intergovernmental effort has produced an explicit definition of "tar" as "all hydrocarbons with molecular weights greater than that of benzene" [13]. There are "tar standard" methods that are developed to take the sample and analysis the tar sample [14,15].

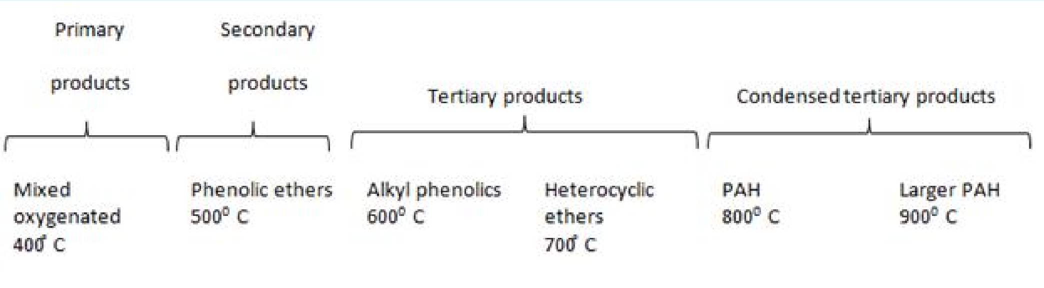


Figure 2: Scheme of tar maturation [6,16].

Based on the reactivity of the compounds, tars can be classified into four product classes: (1) primary products — cellulose-derived, hemicellulose-derived and lignin-derived products. Primary tars form at the pyrolysis step and low temperatures (<500°C); (2) secondary products — phenolics and olefins. In the oxidation step, the primary tars transform and rearrange as secondary tars at a temperature higher than 500°C and reach its maximum at 750°C; (3) alkyl tertiary products — methyl derivatives of aromatic compounds such as methyl acenaphthalene, methyl naphthalene, toluene, indene, phenol and benzene form at temperatures above 650°C and reach to the maximum at ~900 °C; and (4) condensed tertiary products— poly-aromatic hydrocarbons (PAH) series without substituents like benzene, naphthalene, acenaphthalene, antheracene/ phenanthrene and pyrene form at a temperature higher than 750°C [9]. By this token, primary tars are released from devolatilizing feedstock; higher temperatures and longer residence time result in secondary and tertiary tars. Overall, the severe conditions of thermochemical processes produce an array of tarry compounds with diverse properties [16]. Based on the chemical, solubility and condensability of these tar components, tars can be classified into five classes (Table 3) [6,17,18].

Tar class		Representative compounds
Class 1	Heavy tars easily condense at high temperature and very low concentration, GC-undetectable	Determined by subtracting the GC-detectable tar fraction from the total gravimetric tar
Class 2	Heterocyclic aromatic compounds which contain heteroatoms and are highly water- soluble	Pyridine, phenol, cresols, quinoline, isoquinoline, dibenzophenol
Class 3	Light aromatic compounds (1 ring) which do not pose a problem regarding condensability and solubility	Toluene, ethylbenzene, xylenes, styrene
Class 4	Light PAH compounds (2–3 rings) which condense at low temperature even at very low concentration	Indene, naphthalene, methylnaphthalene, biphenyl, acenaphthalene, fluorene, phenanthrene, anthracene
Class 5	Heavy PAH compounds (4–7 rings) which condense at high-temperatures at low concentrations	Fluoranthene, pyrene, chrysene, perylene, coronene

Table 3: Tar classification based on molecular weight [6,17,18].

The tar dew point is as important as the total tar concentration in the application of biomass-derived syngas. Tar dew-point is a powerful parameter to evaluate the performance of gas cleaning systems [19]. Typical tar dew points are between 150°C and 350°C, which is usually far above the lowest process temperature (~30°C). Class 1, 4, and 5 tars influence the tar dew point considerably. They readily condense even at high temperatures and can cause major severe fouling and clogging problems in biomass-derived syngas power systems and efficiency loss. Class 2 and

3 tars include heterocyclic aromatics and benzene/toluene/xylene (BTX) compounds; however, have a low effect on the tar dew point, compete with heavier tars for active sites, and makes problems in the catalytic tar removal method [16]. They are water-soluble, which means they create pollution problems in the aqueous phase of downstream wet gas cleaning equipment [4,20,21].

Tar Removal Methods

There are some sophisticated results available, which have claimed to reduce tar amount significantly. However, the method must be efficient in terms of tar removal, economically feasible, but more importantly, it should not affect the formation of useful gaseous products and leave a less environmental impact. The available methods to reduce tar concentration in syngas down to the desired level can be classified into two types according to the location where tar is removed: primary method (in the gasifier) and secondary method (outside the gasifier) [4,22] (Figure 3). The following sections describe both methods, with emphasis on the secondary method.

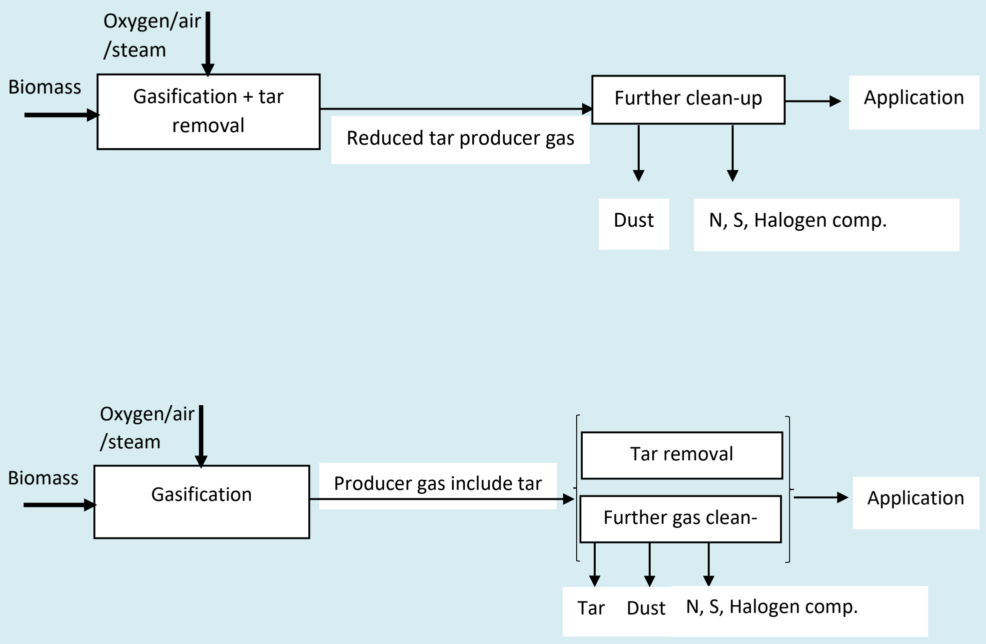


Figure 3: Tar reduction concept by the primary and secondary method [22,23].

Primary Treatments

In biomass gasification, system configuration and operating parameters, such as temperature, pressure, equivalence ratio (ER, O_2 content of air supply/O_2 required for complete combustion), type of feed, gasifying medium, residence time and etc. play key roles in the product distribution, as well as tar generation [24]. In order to remove tar efficiently, numerous techniques related to primary method treatments have been investigated. The current issues of primary treatments are (1) the proper selection of the operating conditions, (2) the use of a proper bed additive or a catalyst during gasification, and (3) a proper gasifier design [22].

Gasifier Operation Conditions:

Temperature

Residence time, temperature, and gasifying agents strongly influence the efficiency of thermal cracking. Lower temperature gasifiers are known to produce excessive tar emissions, which can be reduced through various efficient ways: (1) increasing residence time, such as using a fluidized bed reactor freeboard is partially effective in most cases; (2) using heat exchangers which directly contact tar with an independently heated surface. It required a significant energy supply and decreases the overall efficiency since heat exchangers have the disadvantages of higher energy input and good gas mixing [25] and (3) partial oxidation by adding air or oxygen that could increase CO levels at the expense of a decrease in conversion efficiency and increase the operational cost [4,26].

Increasing the temperature could significantly change the product distribution and reduce the tar content. Tar content, gas heating value, char conversion, and the risk of sintering critically depend on the gasification temperature range [22,27]. In order to get low tar content in the resulting syngas, high operating temperatures (>750°C) are applied to get carbon conversion of the feedstock at a high level. Higher temperatures can lead to the lower yield of tar and higher yields of gaseous products [28,29]. Hence, high-temperature gasifiers are designed to operate at thermal treatment conditions that promote tar decomposition. Temperature not only affects the amount of tar formed but also the composition of tar by influencing the chemical reactions involved in the whole gasification network. Usually, if gasification temperature exceeds 850°C, oxygen-containing compounds such as phenol, cresol, benzofuran, and other 1-ring, and 2-ring aromatics decrease and the formation of Class 4 and 5 (3- and 4-ring) aromatics increase quickly with other unwanted by-products. An increasing temperature also promotes the formation of gaseous products [20,30]. Increasing the temperature inside fluidized-bed gasifiers will reduce tar concentration in the produced gas [22,31]. An intensive cooling system is required to cool down the produced gas in down steam. Ash melts at higher temperatures is another challenge [16,32]. There are several other factors that limit the operating temperature. Tar content, gas heating value, char conversion, and the risk of sintering critically depend on the gasification temperature range [22,27].

Pressure

An increase in pressure could noticeably affect tar concentration and composition. For example, Knight observed at T= 824 ± 8 ^{o}C, steam/feed ratio of 0.76±0.09 (w/w) almost complete removal of phenols for Wisconsin whole tree chips at 21.4 bar compare to 8 bar, but the polycyclic aromatic hydrocarbon (PAH) fraction increased [33]. Wang, et al. reported a decrease in the number of light hydrocarbons (LHC, lower than naphthalene) as well as that of tar in the fuel gas with an increasing equivalent ratio (ER, the required O_2 for gasification to required O_2 for full combustion of the given biomass) for pressurized gasification with 100% carbon conversion [33].

Residence time

Residence time affects either tar yield or tar composition. Residence time has little influence on the tar yield, but it significantly influences the concentration of O_2-containing compounds and 1, 2, 3, 4-ring compounds that depends on the superficial velocity of the wet producer gases in the gasifier. With increasing the residence time, O_2-containing compounds and 1- and 2-ring compounds (except benzene and naphthalene) decreased, whereas that of 3- and 4-ring compounds increased [34]. Corella, et al. observed in biomass gasification with a catalyst, increasing the residence time decreases the total tar content [35].

Gasifying agents

Gasifying agents, such as steam, air, O_2, and CO_2 strongly affect gasification reactions, resulting in different tar compositions and concentrations. Pure steam or mixed steam were used to speed up tar cracking and avoid lower heating value or poor gas composition. The pure steam gasification product is more or less free from N_2 and more than 50% H_2 in the syngas. It was reported SB ratio (steam–biomass ratio (SB); H_2O/biomass [(kg h^{-1}) (kgdaf h^{-1}) $^{-1}$]) affect the gasification products: in spite of the sharp reduction in tar (8% yield at 0.5 SB decreased to almost nil at 2.5), there was a sharp decrease in the lower heating value that was attributed by the decrease in CO [36]. Many researchers used steam–oxygen mixtures for biomass gasification because steam gasification is endothermic and sometimes requires heat supply in the gasifier. Oxygen used as a gasifying medium can provide the necessary heat for gasification, and then the gasifier works as an auto-thermal reactor without complex design. Although the operating parameters and gasifiers are not the same, the researchers report similar results: the effect of gasifier bed temperature on tar content is significant at lower GR values (gasifying ratio: (H_2O + O_2)/biomass [(kg h^{-1}) (kgdaf h^{-1}) $^{-1}$]). Under selected conditions, more tar is formed with pure steam, than that with a steam–O_2 mixture and less with air as gasifying agent [22,37,38]. Although, the results of using gasifying mediums are promising; choosing the proper ratio of steam/biomass and compromising on gas quality is still a big challenge [39,40].

O_2 and air can help to reduce tar concentration. According to the Kinoshita et al. report, tar yield and tar concentration will be reduced when the ER increases because extra oxygen reacts with volatiles in the flaming pyrolysis zone. ER affects both tar concentration and tar composition. At 700 ∘ C, increasing ER from 0.22 to 0.32 reduced tar concentration around 30% but increased PAH fraction in total tar. At ER of 0.27, all phenols are converted but benzene, naphthalene, 3- and 4-ring compounds concentrations increase. The effect of ER can be more considerable at higher temperatures [34]. A similar trend was proved by various experiments with varying ER carried out by other researchers [41,42]. Higher ER value leads to a lower concentration of H_2, CO, and higher CO_2 content in the syngas and decreases the heating value. This effect of ER is more significant at higher temperatures [34].

CO_2, as a promising gasifying medium, also can help with tar reduction. With or without the presence of a catalyst,

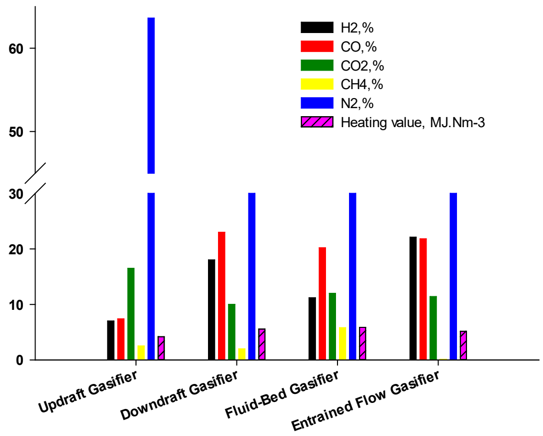
CO_2 can convert into other products. Steam–CO_2 mixture produced the highest activity char, which resulted in high ash content produced gas [40]. CO_2 transforms tar to CO and H_2 in the presence of a catalyst, and reduces the CH_4 and C_2-fraction (C_2H_2; C_2H_4; C_2H_6) [43].

Bed Additives and Catalysts: The 'ideal' bed additives have high efficiency with high selectivity and low cost. Although, catalysts for tar reduction have been extensively investigated, only a few have been tried as active bed additives inside the gasifier itself during gasification. These bed additives act as in situ catalysts, not only influence the gas composition but also affect the heating value of the producer gas. Dolomite attracted more attention among all the bed-active materials. Steam and dry reforming reactions eliminate tar over calcined dolomite. Many researchers have been done using this catalyst to remove tar in gasifiers. It was reported that 1-10 wt% of calcined dolomite is sufficient to improve the quality of the producer gas (reduced 40-80% tar) without changing the heating value since the reduction of CO is compensated by the increase in H_2 production; and CH_4 and C_2H_n concentration do not change considerably in the producer gas [35,44-47]. In addition, using dolomite in proper operating conditions can improve the production of a clean gas significantly. There are other in-bed additives, such as limestone, olivine, Ni-based catalyst, transition metal oxides, potassium carbonate, zeolite in silica–alumina matrix, and char. Bed additives have the following general functionalities [9,22,48]:

1. Change gas distribution,
2. Reduce tar,
3. Increase hydrogen production,
4. Decrease CO slightly and increase CO_2,
5. No variation in the amount of CH_4,

The heterogeneous nature of these additives cause coke formation and deactivation. Attrition and carryover of fines are other problems of these heterogeneous additives which may overload the Particulate Matter removal system downstream of the gasifier. Sometimes adding these low-cost additives could cause the auger jam which is proved to be another major problem to the real-world operation.

Gasifier Design: Reactor design improves the efficiency, heating value of the syngas and reduces tar concentration. Gasifiers are categorized as either fluidized-bed or fixed-bed [49]. Fixed-bed gasifiers produce gas with less particulate content including ash, tar, and char compared to fluidized-bed reactors. Depending on the airflow direction and its feeding point, gasifiers are known as updraft, downdraft and cross-draft (Figure 4) [22,50]. In updraft gasifiers, biomass enters the gasifier from the top, while air is supplied from the bottom. The produced gas in the updraft gasifier contains lots of tar and moisture. In downdraft gasifier, both wood and air are supplied from the top and move downward. The syngas contains a low concentration of tar and particulate. Overall thermal efficiency is relatively low. In cross-flow reactor, the biomass fed on the top and move downward while air enters from the side of the reactor and move upward. So, syngas leaves the gasifier from the top. The produced gas has high tar content and the overall energy efficiency is low [50].



	Updraft Gasifier	Downdraft Gasifier	Fluid-Bed Gasifier	Entrained Flow Gasifier
Reactor temperature, °C	700–900	700–1200	750–950	~1500
Syngas temperature, °C	75–150	850–1000	650–850	1100-1300
Tar, $g.Nm^{-3}$	50–150	0.1–2	1-50	–

Figure 4: Syngas composition at various gasifier configurations [11].

The most popular designs are the secondary or guard bed and the two-stage gasifier. A guard bed can be a fixed, bubbling fluidized, or moving bed that produced syngas pass through that and mixed with a secondary airflow that is injected into the system. The bed can be calcined dolomites, magnesites, and calcites from different quarries and the temperature are 800-900 ˚C [42]. Secondary air injection could increase the temperature which led to a significant tar reduction [42,51]. Secondary to primary air ratio, gasifier size, and temperature are the key parameters for improving the tar removal efficiency. Usually, effective ways are to control secondary to primary air ratio and sufficiently reduce the total tar at certain ranges of gasification temperature; design longer size gasifiers to raise the temperature in the freeboard and bring a higher possibility of lower tar content in the gas.

A two-stage gasifier has been reported to be very effective to remove tar. The gasifier includes the pyrolysis zone and the reduction zone. A secondary air supply is introduced to the second stage (reduction zone) and its temperature is kept as high as possible to crack the tars formed during the first stage (pyrolysis zone). Successful operation of this type of gasifier mainly depends on how stable the pyrolysis zone is [51]. The stabilization of this zone depends on the balance between downward solid movement and upward flame propagation [22,52]. Upward flame propagation should exceed wood consumption to prevent shrinkage of flaming pyrolysis zone and reaching to the second air inlet. Adjustments of the airflow by changing the width of the second air inlet, could keep flame propagation upward. There are several projects running, mainly focused on small scale biomass CHP (Combined Heat and Power) applications and industrial syngas applications Asian Institute of Technology (AIT), Thailand, designed a gasifier which involves two levels of air intakes and could reduce tar about 40 times less than a single-stage reactor under similar operating conditions [51,53-55].

Several attempts have been made to develop two-stage gasification techniques: for example the same reactor design — a combination of pyrolysis of the biomass feed with subsequent partial oxidation of the volatile products in the presence of a charcoal bed was reported by Henriksen, et al. and Brandt et al respectively [52,56]; Susanto, et al. developed a co-current moving bed gasifier with internal recycling and a separate combustion zone for the pyrolysis gas. Their aim was to produce a design suitable for scaling up a downdraft gasifier while maintaining a low tar content in the producer gas [57].

Although most of the attempts made to modify the gasifier design claimed to produce a clean gas, so far the primary treatments are not yet fully understood and have yet to be implemented commercially. These methods involve complex process considerations including the selection of optimized operating parameters, include the effects of active bed materials and the design of biomass feeding and dust removal systems [19]. In fact, there is a reluctance to implement these improved designs on a commercial level. This lack of enthusiasm is also linked to the complex gasifier constructions and/or the decreased gas heating value due to the partial oxidation [23,58].

Secondary Treatments

Various secondary cleaning procedures for different syngas applications can be categorized as physical methods, thermal treatment, and catalytic cracking. Often these treatments take place in the second reactor and they aim to complete tar conversion. It is foreseen complete tar removal is achievable using secondary treatment [4] (Figure 5).

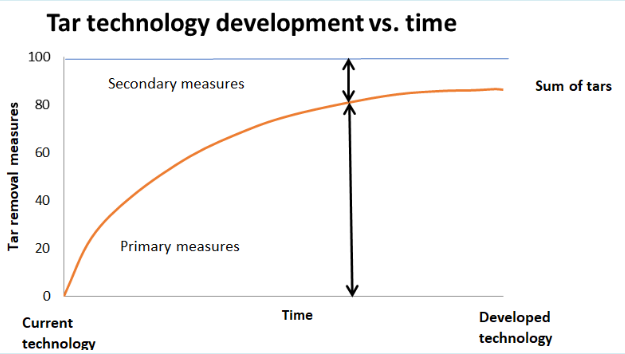


Figure 5: Illustration of the need for primary and secondary measures versus technology development in time [19].

Physical Methods: The physical methods are simple filtration or wet scrubbing of the syngas in order to remove the tar liquid droplets from the gas stream through gas/ solid or gas/ liquid interactions, thus tar removal cannot be separated from the particulate removal. According to the process temperature, it can be classified into two categories: dry gas cleaning and wet gas cleaning. The temperature of dry gas cleaning is usually higher than 500°C and partly below 200°C after gas cooling; while, the temperature of wet gas cleaning is typically about 20–60°C [4,66]. For dry gas cleaning, the devices must consist of temperature tolerable materials, such as ceramics, fiberglass, sand, etc. Cyclone, rotating particle separators (RPS), bag filters, baffle filters, ceramic filters, fabric/tube filters, sand bed filters, adsorbers and etc. are the traditional equipment combined with a gas generation system for tar capturing. Cyclone, rotating particle separators (RPS), or fabric filters alone are not able to reduce the tar significantly, and additional tar reduction may be necessary (Table 4) [23,63,64]; although ceramic filters are considerably efficient in removing tar, at large-scale applications they suffer from their complexity and high cost. Deposition of tar in the filter easily plugs barrier filters [65,67,68]; catalytic filters provide high efficiency in one step [69,70] (Table 5), however, there is not much information related to the investment of catalytic filter in a large-scale system.

Gasifier Type	Feeding Rate (Kg/H)	Gas Cleaning Equipment	Gas Composition (Vol%)					Tar (Mg/Nm3)	LHV (MJ/Nm3)	Cold Gas Eff, (%)	Electrical Eff., (%)
			H_2	CO	CH_4	CO_2	N_2				
Double air stage downdraft [59]	10–12	Cyclone, heat exchangers, and a baghouse filter	16.8	19	0.9	13.6	50.6	<35	4.6	67	
Fixed-bed twin fired [60]	25	A cyclone and a RME (rapemethylester) /H2O quench system followed by a wet electrostatic precipitator(ESP)	18.3	20.4	2.5	14.7	45.5	180-240	5.8	63.5	
Two stage downdraft [61]	45-50	Heat exchanger, bag filter, limestone + activated carbon + desulfurization sorbent packed in a absorber	25– 38	25–38	<2	16-15	10-Aug	20			
Two stage downdraft [62]		Heat exchanger, baghouse filter, paper cartridge filter, demister	32.5	1.5	2.1	19.5	30	15	6		25
Downdraft gasifier	100-110	Cyclone, venturi scrubber, heat exchanger, chiller, mist eliminator, fine filter	24.1	10.7	4.2	4.6	75	10	4.46	75	16
Dual fired downdraft	98	Heat exchanger, baghouse filter, and paper cartridge filter	21.3	20.5	1.1	10.7	89.7	35	5.3	89	21
Dual fluidized bed		Heat exchanger, filter and scrubber	56.6	16.5	12.9	12.3	10	1000			
Two-stage downdraft [62]		Heat exchanger, baghouse filter, paper cartridge filter, demister	32.5	1.5	2.1	19.5	30	15	6		25
Downdraft gasifier	100-110	Cyclone, venturi scrubber, heat exchanger, chiller, mist eliminator, fine filter	24.1	10.7	4.2	4.6	75	10	4.46	75	16
Dual fired downdraft	98	Heat exchanger, baghouse filter, and paper cartridge filter	21.3	20.5	1.1	10.7	89.7	35	5.3	89	21
Dual fluidized bed		Heat exchanger, filter, and scrubber	56.6	16.5	12.9	12.3	10	1000			

Table 4: The details of tar removal from cold gas cleaning including the type of gasifier, type of feedstock, use cleaning equipment, and produced gas composition and quality [58].

Equipment	Tar reduction	Key consideration
Cyclone	50–90%	A humidifier/cyclone combination removed coal tar, no data on efficiency was found for biomass tar removal; Effective to large particulate compared to tar removal
Rotating particle separators (RPS)	30–70%	Insufficient tar reduction and additional tar reduction is necessary; The device is more applicable for solid particulates than tar removal
Ceramic filters	75-98%	Complexity and high investment; Plugging
Fabric/tube filters	0–50%	Insufficient tar reduction and additional tar reduction is necessary; Plugging
Sand bed filters	50–97%	Plugging
Catalytic filter	>90%	High cost, although particles removal and tar catalytic cracking happen in one step (96% and 98% for naphthalene and 41% and 79% for benzene); Plugging; Coking deposition; Poisoning
Absorbers	>70%	Consumable; Issues from absorbers regeneration or disposal

Table 5: Tar reduction in dry gas cleaning systems [4,23,63-66].

Compared to dry gas cleaning, wet gas cleaning demonstrates good performance to remove particles and condensable tar droplets from producer gas both on the laboratory and industrial scale and commonly used cold gas cleaning methods. Scrubbers (packed column, spray tower, impingement, and venturi), wet electrostatic precipitators (ESP), OLGA, wet cyclones are common wet scrubbers. Water, used and fresh, vegetable oil, and biodiesel are common scrubber media [71,58]. If the water is used as a scrubber media, water scrubbing produces large amounts of toxic waste which brings the excessive cost related to wastewater treatment and sludge disposal [72]. Water scrubbing can cool down the syngas and captures the solid particles and/or liquid drops, and also water-soluble gases such as NH_3, HCl, H_2S, and SO_2. In a large-scale gasification system, water scrubbing needs a remarkably large size scrubber with a large amount of water supply to provide enough residence time for impure gas dissolution in the scrubber which will increase the cost significantly. In this case some of the toxic gas escapes from the scrubber, an additional system is required for further gas removal.

Since the producer gas temperature of the pilot-scale gasifier is around 800-950°C, cold gas cleaning usually happens after cooling. Bhave and Patel, 2008 designed a packed-bed scrubber that included wet and dry sections [73]. Rasching ring and stones used for packing and water was the scrubber media. The system was able to reduce the tar and particle concentration to less than 150 mg/Nm^3. After a certain stage, the sand beds need to be washed with detergent solution, dried, and sieved before they can be reused [74]. The Güssing system installed in Austria used organic solvent scrubbers to remove tar, ammonia, and acidic gas components. They used organic solvent as the scrubbing agent that was partially replaced by the fresh agent continuously. The contaminated agent was then recycled to the combustion zone of the gasifier. So, No residue or wastewater accumulated during the gas cleaning and cooling steps (Table 6 & 7) [68]. Table 8 summarized the details of cold gas cleaning for syngas generated from pilot-scale biomass gasifiers, including the feedstock type, process details, gas quality, and some other concerns. Table 9 demonstrates the applications of different scrubbers based on the contaminants. Venturi scrubbers have higher tar removal efficiency (up to 90%) compared to other scrubbers [74]. Wet electrostatic precipitators have proved that they have good tar removal efficiency; however, cost efficiency limits their application to small-scale operations [73]. Although wet scrubbing is an effective way to reduce tar concentration, the significant disadvantages cannot be neglected (Table 9) especially since the process requires significant heating and cooling steps unless the gas is needed at near-ambient temperature. These heat penalties decrease the overall process efficiency including low carbon conversion and low gas efficiency [75]; low burnable gas composition [60]; uneconomical [76] and the most serious issue is that a large amount of toxic waste residue, such as wastewater is produced which requires downstream treatment and/or recycles, and has big environmental impacts and boosts the operation cost (Table 6, Table 9). Up to date, researchers still keep looking for constructive solutions such as using novel scrubber emulsions as washing agents and developing the new gas cleaning system to lower the operation cost, increase the performance efficiency, and leave less environmental impact [77,78]. Storage of the produced gas provides flexibility in its usage and design of gasifier and could improve the economic viability of the gasifier [79] by using it in a proper place and time or conversion to chemicals [80]. Ahmed, et al. compress the gas up to 0.86MPa to store the produced gas [81]. Gas compression remove tar, as well. Din, et al. first passed the producer gas through an oil scrubber and charcoal and reduced the tar concentration to 138–312 ± 31 mg Nm^{-3}. Then, the gas was introduced into a compressor. Compressing of gas to 0.8 MPag condensed tar inside the compressor receiver tank and reduced tar concentration to 84.4± 1.2% [80].

Name	Gas cleaning process (GCP)			Waste water treatment	Waste water recycling
	Dry GCP	Wet GCP	Process detail		
Güssing	✓	✓	Tube filter and wet tar washing	Evaporation	Combustion of residues in the plant
Harboore		✓	Quench and wet ESP	Sedimentation, evaporation	Combustion of residues in the plant
Wiener Neustadt		✓	Quench and wet ESP	Evaporation	Disposal of residues
Pyroforce	✓	✓	Tube filter and wet tar washing	Storage and disposal	Disposal of residues or utilization in the process
IWT test facility / shaft gasifier	✓	✓	Tube filter and wet tar washing	Staged waste water treatment, evaporation, vapor stripping and residue recycling	Recycling in the process, discharge of waste water into the sewer system possible
DTU test facility / multi-stage gasifier	✓		Dry gas de-dusting with tube filter	Unnecessary	Recycling in the process
IWT test facility / multi-stage gasifier	✓		Dry gas de-dusting with tube filter	Unnecessary	Recycling in the process

Table 6: Various concepts of wastewater treatment related to cold gas cleaning [4].

Recently, OLGA that has been developed by the energy research center of the Netherland (ECN) demonstrated on the 0.8 MWt ECN MILENA pilot facilities, in France on a 4 MWt facility, and in Portugal on the 4 MWt Tondela facilities. The basic principle of OLGA is dew point control (Figure 6). In the first OLGA loop, a scrubbing oil cools down the gas gently (350˚ C to 150˚ C), and condensate heavy tar particles. OLGA prevents water condensation and its mixing with tar [78]. The collected tar is separated from the scrubbing oil and is recycled back to the gasifier. In the second loop that is called the absorber column, the scrubbing oil absorbs lighter tars at lower temperatures (< 60˚ C) [82]. The saturated oil passes through a stripper. Hot air or steam is used to regenerate the oil. Recycling all the heavy and light tars to the gasifier will break down them, improves the energy efficiency, and eliminate tar waste stream [83]. OLGA removes 99% phenolic and 97% heterocyclic compounds. These advanced technologies may be suitable for some applications such as fuel cells and chemical synthesis like Fischer–Tropsch. However, concerning high production cost or high energy requirements, these advanced tar removal technologies may affect the cost feasibility [84,85].

Feedstock type	**Feeding rate (kg/h)**	**Gas cleaning process (GCP)**			**Tar (mg/ Nm^3)**	**LHV^3 (MJ/ Nm^3)**	**Cold gas eff., (%)**	**Electrical eff., (%)**	**Note**
		Dry	**Wet**	**Process details**					
Eucalyptus wood [86]	10–12	√		Cyclone, heat exchangers, and a baghouse filter	<35	4.6	67		Cold gas efficiency is low. The tar concentration in the final clean gas was much lower than the acceptable range for an IC engine. It increased the cleaning cost.
Woodchips [87]	25	√	√	A cyclone and a RME (rapemethylester) /H2O quench system followed by a wet electrostatic precipitator(ESP)	180-240	5.8	63.5		Water sprayed to quench the syngas to 50°C, then a RME /H_2O was used. The tar content doesn't meet specification of IC engine. The solvent are expensive
Corn cob [60]	45-50	√		Heat exchanger, bag filter, limestone + activated carbon + desulfurization sorbent packed in a absorber	20				
Woodchips [56]		√		Heat exchanger, baghouse filter, paper cartridge filter, demister	15	6		25	The bag house filter was an excellent well operating gas cleaning system, based on 465 h testing
Olive Kernel [59]	100-110	√	√	Cyclone, venture scrubber, heat exchanger, chiller, mist eliminator, fine filter	10	4.46	75	16	The heating value and the overall electrical efficiency were low

Wood [88]	98	√		Heat exchanger, baghouse filter and paper cartridge filter	35	5.3	89	21	The cold gas efficiency was quite high, while the gas heating value was almost the same. This indicates that the energy input for both in the gasification section and gas cleaning section is quite low
Woodchips [89]		√	√	Heat exchanger, filter and scrubber	1000				The gas yield is 1.29 Nm^3/kg, which is lower compared to air blown gasifier. The heating value is much higher than that of air gasification
Woodchips [75]	250	√	√	Cyclone, spray tower, packed column scrubber, condenser, a purification tower, two wire mesh mist eliminator		4.7	53	16	The carbon conversion (73.91wt%) and cold gas efficiency are low. The gas yield was 2Nm^3/kg
Sunflower seed and sawdust [90]	54	√	√	Cyclone, venture scrubber, chiller condenser, two sawdust filter and a bag filter		5.6	67.7		The purification tower and condenser were optimized for tar removal
Poplar woodchips [86]	57-92	√		Pilot gas cleaning using CaO absorber, cyclone and cold gas filter		2.1	12.7	70	fluidized bed pilot gasifiers
Sewage Sludge [87]	570-1140	√	√	Cyclone, gas cooler, granular bed filter, Ceramic filter, water absorber, packed column for NH_3 and H_2S	<100		4.7	70	The burnable gas composition and the heating value was too low

Table 7: Summary of the details of cold gas cleaning for syngas generated from pilot-scale biomass gasifier [58].

No.	Method	Application
Low energy scrubbers	Spray scrubbers	To remove coarse particulate matter, and remove gaseous pollutants moderatly
	Wet cyclonic scrubbers	Moderate/high removal of particulate matter (< 5μm)
	Vertical baffled scrubbers	Moderate/high removal of coarse particulate matter (< 3μm) & gaseous pollutants
	Horizontal baffled spray scrubbers	High-efficiency for gaseous pollutants
	Packed bed scrubbers	High-efficiency for gaseous pollutants
High energy scrubbers	Venturi scrubbers	Very high-efficiency for fine particulate with partial removal of gaseous pollutants.
	Ejector scrubbers	Very high-efficiency for coarse particulate and gaseous pollutants
	High efficiency wet filters	Very high-efficiency for fine particulate matter soluble in scrubbing liquid
	Cleanable high efficiency air filter	To remove aerosols P_2O_5, SO_3, etc.
	High efficiency air filter	To remove sticky and oily particulates, aerosols and mists

Table 8: Scrubbers and their applications [91].

Equipment	Description	Tar reduction		Key consideration
Wet scrubber	Using water to scrub condense the tar from syngas and remove the particulates	Packed column scrubber(spray tower)	10–25%	Poor regeneration efficiency and continuous operation behaviour. Crucial disadvantages: • Saponification; • Low solubility of hydrocarbon compounds; • Surface tension effects; • Clogging of apparatus; • High costs; • Lower heating value of the syngas and the net energy efficiency of the process for treatment or disposal of wastewater and sludge;
		Venturi scrubber	50–90%	
		Packed bed scrubber	<75%	
		Impingement scrubber	70%	
Wet ESP	The wet ESP process involves a corona discharge producing ionized gas passing between a high voltage electrode and an earthed (grounded) electrode. The ions attach themselves to the dust particles or droplets of tar and the water when charged are attracted to the grounded electrode due to the electric field.	40-70%		Large size and high capital cost making it not suitable for small-scale operations
OLGA	A two-loop oil scrubbing system	99% phenol and 97% heterocyclic tar removal		High production cost
	1.Removing heavy tars and dust in the 1st loop as a liquid slurry 2.Removing light tars such as naphthalene and phenol in the 2nd loop dissolved in air or steam			

Table 9: Tar reduction in wet gas cleaning systems [4,92,93].

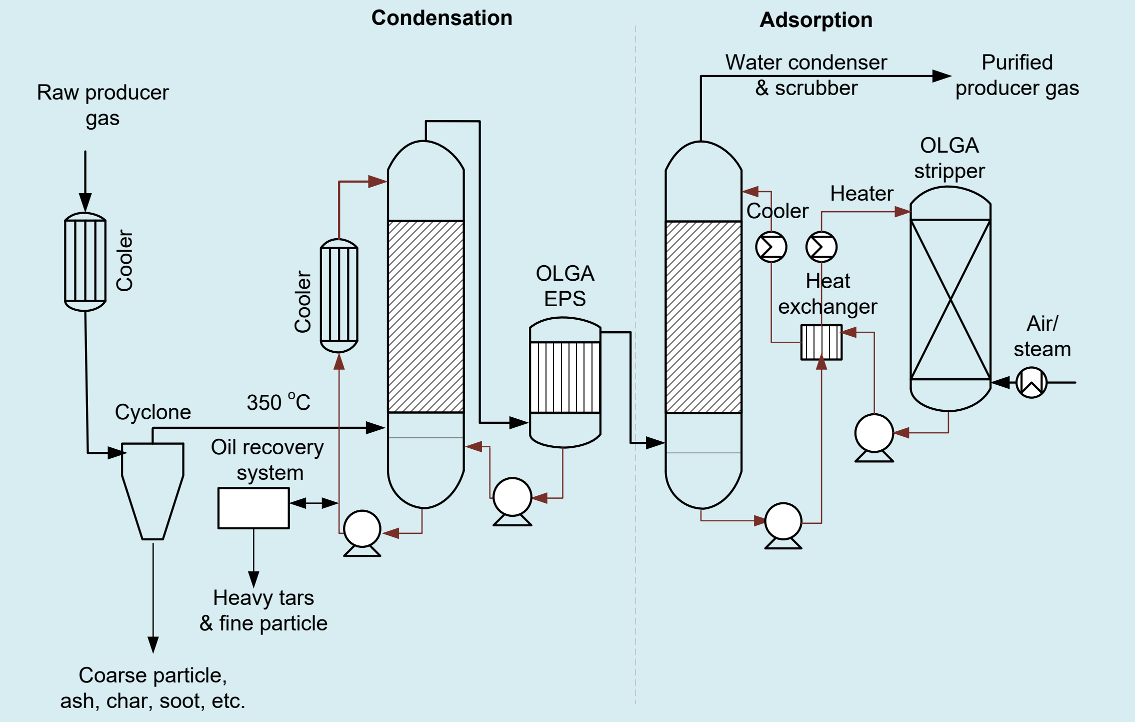


Figure 6: Flow sheet of OLGA [83].

Thermal treatment: Hot gas cleaning, removing the impurities under hot conditions usually at gasification temperature, is one of the more efficient but challenging steps to make syngas which is generated from the pilot biomass gasifiers meet the application requirement. Depending on the types of impurities, hot gas cleaning can be classified either by physical filtration or chemical cracking. Hot gas cleaning could be thermally more efficient rather than cold gas cleaning depends on the downstream application. In cold gas cleaning, first gas is cooled and cleaned and then is heated up to the desired temperature [58,94].

Thermal cracking

Thermal cracking outside gasifier is a heating process that aims to eliminate tar. Temperature and residence time are two key parameters for thermal cracking. Tar is converted or cracked into lighter and less problematic smaller molecules such as methane, carbon monoxide, and hydrogen. The typical temperatures for thermal cracking without involving catalysts range from 1000°C to 1300°C. Some studies show the tar cracking usually happens at temperatures between 700°C and 1250°C [95,96] (Table 10). The residence time required for effective cracking depends on the temperature employed. Gasification of various biomasses such as wood, coconut shell, and straw demonstrated thermal cracking of tar is effective above 700°C [97]; Brandt claimed that the necessary temperature and residence time for tar decomposition were 1250°C and 0.5s, respectively [62,98,24]. Fassinou, et al. reported that increasing the temperature and the residence time improve tar cracking, gas production, and char quality (fixed carbon rate more than 90%, volatile matter rate less than 4%) from pyrolysis of Pinus pinaster in a two-stage gasifier [28]. Thermal cracking of tar from different raw materials at three different temperatures, 700, 850, and 900 ◦ C showed the amounts of benzene and naphthalene increase with temperature, while other light tar compounds decrease [99]. Houben, et al. studied thermal tar treatment, naphthalene as a tar model compound, via partial oxidation. The total tar content slightly decreases at higher air/fuel ratio values, while the tar content strongly decreases at lower air/fuel ratio and higher hydrogen concentrations. It was found that the partial combustion burner reduces the tar content of the gas by over 90% by cracking with an air/fuel ratio of 0.2; the maximal tar reduction reached 98–99% at 900 °C with an excess air ratio of 0.5 [100,101]. Hoeven, et al. reported that rising fuel hydrogen content is always beneficial to improve tar cracking, by increasing reaction rates, radical production, and radical residence times [102]. Minimum tar content obtained at 900°C with an excess air ratio of 0.5, while Brandt and Henriksen partially oxidized pyrolysis gas in a reactor at 800, 900 and 1000° C. Excess air ratio was varied from 0 (thermal cracking) to 0.7 [95].

Thermal cracking may also produce soot that reduces the energy content of the syngas and is not suitable for many stringent applications such as fuel cells and synthetic chemicals. Also, it increases the particulate load on cleanup or processing equipment. Although so much effort has been made for tar removal, no thermal cracking can be achieved in one step with high efficiency and decent cost, as Bridgwater mentioned that biomass-derived tar is very hard to crack by thermal cracking alone [66].

Recent development in the pilot-scale biomass gasification process shows the most thermal cracking happens in the partial combustion zone or so-called the oxidation zone inside the gasifier. During the thermal cracking, a higher O_2–fuel ratio is kept for maintaining higher temperatures at the oxidation zone and effectively removes the tar content in the syngas; however, it reduces the gas heating value due to the reduction of burnable gas composition in the syngas [58]. The downdraft gasifier commonly benefits thermal tar cracking because it can easily maintain the high temperatures required. The concentration of H_2, CO, and CH_4 normally varies, which results in lower and variable heating values between 3 to 6MJ/Nm^3. Meanwhile, the tar content in the syngas varied widely. Although the gas composition is quite enough to run the internal combustion engine, the tar content is much higher in most of the reported work than the acceptable range [58].

Gasifier	Feeding rate (kg/h)	Temperature (°C)	Gas composition (vol%)				Tar (mg/ Nm^3)	LHV (MJ/ Nm^3)	Cold gas eff., (%)
			H_2	CO	CH_4	CO_2			
Downdraft [103]	12	1000	14	24	2	14	<50	5.8	60-78
Downdraft [104]	18.7	1000	8.7-13.2	20.8-23.6	3.6-5.2	9.3-14.5	4800	6.1	67
Updraft and Downdraft	30	900	10.4	15.1	0.3	12.8	450	3.2	
Regenerative [105]	5	1000	14.1-16.3	14.2-21.6	5.2-2.5	15.2-10.3	44-107	5.2-5.4	
Downdraft [106]									
Downdraft [107]	Pilot	954	11.1-20.9	14.3-20.2	2.9-2.8	-	45	4.2-6.0	60
Downdraft [108]	5.4	1000-1200	11.1	18.6	2.2	11.2	3000	4.7	
Downdraft [109]	4-Mar	1050	11.1	18.6	2	13.12	5	3.8-4.0	63

Table 10: Details of thermal cracking data derived from different pilot-scale gasifiers [58].

Plasma cracking

Plasma cracking of tars is a relative of reforming methods. Plasma contains free radicals, ions, and excited molecules which carry enough energy and create a highly reactive atmosphere to initiate tar decomposition reactions [110,111]. Plasma cracking has been widely investigated for pollution control and is classified as either thermal or non-thermal, based on the plasma temperatures [20]. Thermal plasmas are applied in single-stage systems — mainly serving as a heat source and two-stage systems — refining the raw syngas upon exiting the gasification. Tetronics develop a two-stage plasma-arc system combined with a gasification reactor to clean the resulting syngas [112]. Usually, residual tars and chars converts at high-temperature environment in the plasma conversion reactor, and ash are vitrified into a non-leaching slag. Non-thermal plasma systems include pulsed corona, dielectric barrier discharges, DC corona discharges, and radio frequency (RF) plasma, and microwave plasma. A numerical study of naphthalene removal by non-thermal plasma was undertaken using pulsed corona plasma. Tar model compounds in synthetic gas mixtures were successfully removed at different temperatures. The naphthalene conversion is about 90–95% [113-115]. Van Heesch, et al. determined that pulsed corona discharges can crack heavy tars and found the reaction of naphthalene with excited nitrogen molecules plays a key role in the cleaning process [116,117]. ECN has performed research on glide-arc plasmas. This type of plasma lies somewhere in between thermal and non-thermal plasma: 20% of the energy is dissipated in the thermal part, 80% in the 'cold' part [118]. Although the energy density of the plasma was 25% of the producer gas energy content, tar removal efficiency was less than 50% at 600°C [63]. Furthermore, the gliding arc reactor did not show any selectivity towards specific hydrocarbons, not even at elevated temperatures [118].

Disadvantages such as a limited lifetime of the pulsed power devices, high costs, and high energy demand of the overall process strongly limit plasma real-life applications; Although, they are relatively effective for tar removal from syngas [119].

Catalytic cracking: Catalytic cracking is a technically and economically attractive approach for syngas cleaning. Catalysts can reduce the activation energy for tar decomposition, increase tar elimination reaction rates and make reactions thermodynamically feasible at lower temperatures. For the secondary method, tars are converted in a separate reactor. It has the potential to avoid the thermal penalties and costs associated with higher temperature operation. The chemistry involved in catalytic tar decomposition of syngas is a complex mix of hydrocarbon decomposition and equilibrium reactions (Table 11) [8]. The catalysts are divided into three primary components: (1) an active catalytic phase or metal; (2) a promoter, to increases activity and/or stability; and (3) a high surface area support that facilitates dispersion of the active phase [74]. Sutton and co-workers [48] summarised the criteria for catalysts as follows: (1) the catalysts must be effective in removing tar, (2) if the desired product is pure syngas (H_2 and CO), the catalysts must be capable of reforming methane; (3) the catalysts should provide a suitable H_2/CO ratio for the intended process; (4) the catalysts should be resistant to deactivation as a result of carbon fouling and sintering; (5) the catalysts should be easily regenerated; (6) the catalysts should be strong [18] (Table 12).

Reaction	Equation	
Drying and Pyrolysis		
Drying	$H_2O_l \xrightarrow{k_d} H_2O_g$	[120], [121]
P1	$X \xrightarrow{k_{p1}} v_c CHAR + v_{T1} TAR1 + v_{co} CO + v_{co_2} CO_2 + v_{H_2} H_2 + v_{CH_4} CH_4 + v_{H_2O} H_2O$	[121]
P2	$TAR1 \xrightarrow{k_{p2}} v_{T2'} TAR2 + v_{co'} CO + v_{co_2'} CO_2 + v_{H_2'} H_2 + v_{CH_4'} CH_4$	[121]
Combustion		
C1	$(T1) CH_{1.522} O_{0.0228} + 0.867 O_2 \rightarrow CO + 0.761 H_2O$	[122]
C2	$CH_4 + 1.5 O_2 \rightarrow CO + 2H_2O$	[123]
C3	$2CO + O_2 \rightarrow 2CO_2$	[121]
C4	$2H_2 + O_2 \rightarrow 2H_2O$	[121]
C5	$(T2) CH_{1.522} O_{0.0228} + 0.867 O_2 \rightarrow CO + 0.761 H_2O$	[121]
C6	$CH_\alpha O_\beta + \gamma O_2 \rightarrow \left(2 - 2\gamma - \beta + \frac{\alpha}{2}\right) CO + \left(2\gamma + \beta - \frac{\alpha}{2} - 1\right) CO_2 + \frac{\alpha}{2} H_2O$	[122][124]
Gasification		
G1	$CH_\alpha O_\beta + CO_2 \rightarrow 2CO + \beta H_2O + \left(\frac{\alpha}{2} - \beta\right) H_2$	[122][124]
G2	$CH_\alpha O_\beta + \left(2 - \frac{\alpha}{2} + \beta\right) H_2 \rightarrow CH_4 + \beta H_2O$	[122][124]

G3	$CH_\alpha O_\beta+(1-\beta)H_2O\rightarrow CO+\left(1-\beta+\frac{\alpha}{2}\right)H_2$	[122][124]
Reduction		
Steam reforming1	$CH_4+H_2O\rightarrow CO+3H_2$	[122]
Steam reforming1	$(T2)CH_{1.522}O_{0.0228}+0.978H_2O\rightarrow CO_2+1.74H_2$	[122]
Gas-water shift	$CO+H_2O\rightarrow CO_2+H_2$b	[121]
Possible tar decomposition reaction		
Steam reforming	$Tar^*+nH_2O\rightarrow nCO+(n+0.5m)H_2$	[8]
Steam dealkylation	$Tar^*+xH_2O\rightarrow C_xH_y+qCO+pH_2$	
Thermal cracking	$Tar^*\rightarrow C^*+C_xH_y+gas$	
Hydro cracking	$Tar^*+(2n-(m/2))H_2\rightarrow nCH_4$	
Hydro dealkylation	$Tar^*+xH_2\rightarrow C_xH_y+qCO+qCH_4$	
Dry reforming	$Tar^*+nCO_2\rightarrow 2nCO+0.5mH_2$	
Cracking	$C_nH_{2n+2}\rightarrow C_{n-1}H_{2(n-1)}+CH_4$	
Carbon formation	$C_nH_{2n+2}\rightarrow nC+(n+1)H_2$	
Combustion reaction		
Water-gas shift	$CO+H_2O\rightarrow H_2+CO_2$	[8]
Methanation 1	$CO+3H_2\rightarrow CH_4+H_2O$	
Methanation 2	$2H_2+C\rightarrow CH_4$	
Water gas shift	$CO+H_2\rightarrow H_2O+C$	
	$CO_2+2H_2\rightarrow 2H_2O+C$	
Boudouard	$C+CO_2\rightarrow 2CO$	

Table 11: Important decomposition and equilibrium reactions of gasification and tar removal

*C_nH_m ; $CH_\alpha O_\beta$ *represent char*

Mineral	Clay minerals	
	Calcined rocks	• Calcite • Magnesite • Calcined dolomite • Olivine
	Ferreous metal oxides	
Synthetic catalysts	Char	
	FCC catalysts	
	Transition metals based	• Ni-based • (Pt, Zr, Rh, Ru, Fe)-based
	Activated ammonia	
	Alkali metal carbonates	

Table 12: Classification and types of catalysts used for tar elimination [18].

The catalytic cracking mechanisms have been investigated extensively and many excellent reviews also provide in-depth information related to the different catalysts composition and conditioning [4,8,16,59,125]. K-enriched Ca-oxide formed as an outer layer on olivine particles is able to reduce tar content in produced gas [126]. Ni-based catalysts are able to reform heavy hydrocarbons such as tar in the presence of steam but they suffer from fast deactivation causing by carbon deposition and sintering. Sulphur poisoned them easily. Mayenite ($Ca_{12}Al_{14}O_{33}$) as support is able to reduce deactivation rate but still has no effect on sulphur poisoning. Savuto, et al. 2018 demonstrated adding CeO_2 to Ni/Mayenite catalysts reduced carbon deposition [127]. Carbonaceous materials such as tyre char demonstrate high activity in tar removal from the produces gas [128]. Active carbons reduce the tar and condensable gas concentration. Impregnation of active carbon by Ni increases the H_2 content up to 26 vol% and reduces NH_3 concentration to 198 ppmv. Fe-impregnated active carbon deducts H_2S concentration to 96 ppmv. Their surface area and porosity diminish versus time [129]. Tables 13 & 14 show some of these catalysts and their highlighted characteristics.

Catalysts		Chemical composition related to catalytic activity	Factors of catalytic activity	Factors of catalytic deactivation	Advantages	Disadvantages	Some important results
	Calcined rocks (limestone, magnesium carbonate, dolomite and olivine)	CaO and/ or MgO with different concentration of Fe_2O_3	Effective pore size Calcination (increase surface area and lead to iron species migration on the surface) Relatively high alkaline (K, Na) content.	Coke deposition CO_2 partial pressure	Inexpensive and abundant The most popular cheap catalysts	They are very soft and quickly eroded in fluidized beds with high turbulence Olivine has higher attrition resistance	The activity can be improved by increasing the Ca/Mg ratio, decreasing the grain size, and increasing the active metal content such as iron. The reactivity: calcined dolomite > calcite > magnesite. In situ use of dolomite is less effective than its use downstream from the gasifier

Minerals	**Clay minerals.** (kaolinite, montmorillonite, and illite)	Al_2O_3 and/ or SiO_2	Effective pore size Internal surface area number of acidic sites	Temperature >800°C	Relatively Inexpensive Less disposal problems	Lower activity compared to dolomite	Commercial nickel catalyst (Ni on Al_2O_3) > dolomite > activated alumina > silica-alumina (clay mineral) > silicon carbide (inert)
	Iron Ores	Iron oxides, or carbonates, or sulfides, or silicates	H_2 concentration	Coke deposition	Relatively Inexpensive	Lower activity compared to dolomite	Iron oxide-silica in its reduced form has a high activity toward benzene cracking and a high selectivity toward methane formation.
Synthetic Catalysts	**Transition metal-based Catalysts**	Ni, Rh, Ru, Pd, Pt, etc	Temperature Promoter(s) type and content Support type, and the content (acidity, surface area, pore structure, and electronic structure)	Mechanical deactivation (attrition) Sintering (high temperatures). Coke formation Poisoning (mainly H2S)	Good catalysts for the steam and dry reforming of methane and hydrocarbons Ni-based catalysts are 8-10 times more active than dolomite	Rapid deactivation because of sulfur and high tar content in the feed Relatively expensive Secondary pollution	Promoters are usually transition metal-based catalysts such as Mo, W, Zr, Mn, lanthanides such as La and Ce, and Al, which increases activity and/or stability Support gives a high surface area, durability, and coking resistance Al_2O_3 is the most common support Order of activity to be Rh > Pt > Ni > Pd > Ru
	Char	C with mineral content	Effective pore size Internal surface area The ash or mineral content of the char.	Coke deposition Catalyst loss	Relatively Inexpensive Natural production inside the gasifier Higher tar conversion compared to dolomite	Need continuous external supply	The reactivity of char produced in the pyrolysis stage is highly affected by the treatment conditions, and they thought it might significantly increase if high heating rates, small fuel particle sizes, and short residence times at high temperatures were used.

	Fluid catalytic cracking (FCC)	Zeolites (frameworks of $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$ coordination polyhedra)	Si/Al ratio Particle size Nature of the (exchanged) cation	Coke deposition Catalyst poisoning (steam, basic nitrogen compounds, and alkaline metals)	Relatively Inexpensive Thermal/ hydrothermal stability Better resistance to nitrogen and sulfur compounds Tendency toward low coke formation Easy regenerability.	Rapid deactivation because of coke formation	The composition of coke strongly depends on the cracking temperature and that the H/C ratio decreases with increasing temperature.
	Alkali-metal-based catalysts	Any of the monovalent metals Li, Na, K, Rb, Cs, and Fr, belonging to group 1A of the periodic table.	Alkali metal species	>900°C melting and agglomeration	Natural production in the gasifier reduce ash-handling problems	Particle agglomeration at high temperatures Lower catalytic activity than dolomite	Alkali metal carbonate used and has the order Na ≥ K > Cs > Li. Catalytic activity of single salts in steam gasification depends on the gasification temperature, with the following order of activity: K2CO3 > Ni(NO3)2 > K2SO4 > Ba(NO3)2 > FeSO4.
	Activated alumina	$Al_2O(_{3-x})(OH)_{2x}$	Complex mixture of aluminum, oxygen, and hydroxyl ions that combine in specific ways to produce both acid and base sites	Coke formation.	High tar conversion comparable to that of dolomite	Rapid deactivation because of coke formation	Activated alumina was nearly as effective as dolomite.

Table 13: Summary of catalysts used for tar elimination [18,130].

Catalyst	tar	Process	T(˚C)	P (atm)	Efficiency	Comment
Ni-Fe-Mg [131]	Toluene & ethylene	Steam reforming-Fixed-bed reactor	600-800	1	~100%	No coke formation for 48h
Activated carbon, Ni-impregnated active carbon, Fe-impregnated active carbon [129]	Produced from Gasification of dried sewage sludge	^{01}Tar cracking reactor- Fixed-bed reactor (UOS[1] three-stage gasification process)	650-815	1	67%-92%	Air/fuel~0.3 t~1s
Rice husk char (RHC) and metal impregnated (Fe, Cu and K) char [132]	Tars from biomass pyrolysis	Dual-stage reactor, Fixed-bed reactor	800	1	77.1%-92.6%	Increase H_2, CO and CH_4 yield
Char [133]	Tar from fluidized bed two-stage (FBTS) gasification process	Fluidized-bed reactor	1000	1	75%	t = 0.9s
Char supported nickel catalyst [134]	Tar of steam gasification	Lab-scale fixed bed reactor	800	1	NA	t =0.5s Reduced heavy polyaromatic hydrocarbons in tar
Waste-derived char and ash (metal oxides) [135]	Solid residues from a commercial gasification plant	Fixed-bed	900	1	85%,	Reduction of tar from 65 g/Nm^3 to 90 and 173 mg/Nm^3
Dolomites and Ni-based [136]	Toluene	Fixed-bed- steam reforming	700-800		~100%	Coke formation at low temperature even at high steam/ carbon ratios
Ni and Mn perovskite-based catalysts [137]	Toluene	Fixed-bed- steam reforming	700			Higher activity and stability in the absence of H_2S
Calcined scallop shell supported Cu catalysts [138]	Cedar wood gasification	Fixed-bed- steam reforming	600-750	1		Calcium copper oxide phase could stabilize Cu species. Strong basicity of CS support enhanced the Cu/CS activity.

Table 14: Developed catalysts for tar removal.

The catalytic gas upgrading reactor is the key tar removal equipment in CHRISGAS (Clean Hydrogen-rich Synthesis Gas) project that was launched the first plant to develop highly efficient and environmentally acceptable technologies based on biomass that was built in Sweden in 1990s [139]. In the new CHRISGAS syngas process, dry or pre-dried material and steam/oxygen enter a fluidized bed; the producer gas passes through a hot gas filter to remove the particulate; tars are reforming in a catalytic gas upgrading reactor, i.e. steam reformer; and the water-gas shift and hydrogenation reactors. The raw synthesis gas are processed further for conversion to a vehicle fuel (Figure 7) [139]. A number of successful tests which ran in the scale of 4000kg/h biomass feeding were conducted at the IGCC plant [140]. Different catalysts for tar reforming and catalyst for high temperature water–gas shift reaction were comprehensively tested for H_2 rich clean gas production [104,141,142]. Interestingly, some catalytic tar cracking results under CHRISGAS did not support the conclusions of the investigation made based on the bench scale reactions. For example, olivine-magnesium iron silicate $(MgFe)_2SiO_4$ did not provide significant impact in both reduction of tar and elevation of hydrogen in the syngas [143-145]; thermally pretreated (calcined olivine) and untreated catalysts provided almost similar results as the base case (using sand) in terms of gas composition and tar reduction [146]. In contrast, magnesite ($MgCO_3$) exhibited excellent tar reforming activities, provided almost double of H_2 in the producer gas [146]. In the CHRISGAS project, using magnesite in the primary bed and Ni–$MgAl_2O_4$ in the secondary reformer, the tar reduction was achieved to the acceptable level for downstream application; however, the sulfur poisoning of nickel catalyst is a big drawback [147]. Under the CHRISGAS project, it was also proved that Cu-based sorbent were better than the Ca and Fe based sorbents and provided excellent results for H_2S removal; and bauxite and kaolinite were suitable sorbents to reduce KCl concentration [147].

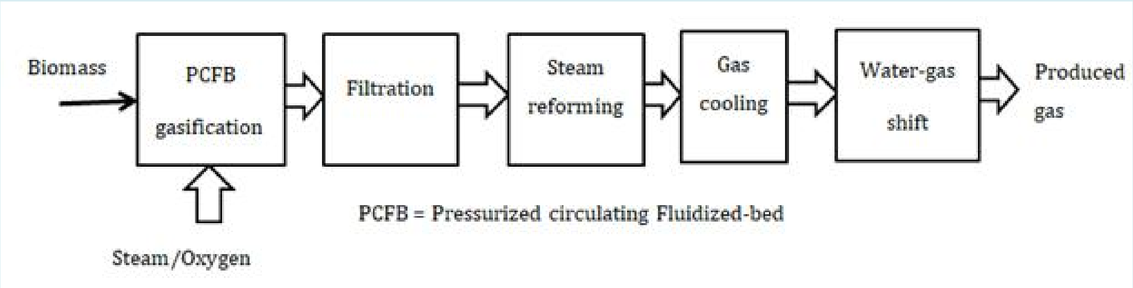


Figure 7: Block diagram of the CHRISGAS syngas process [140].

Other research groups have also done extensive studies focusing on catalytic tar cracking at demonstration and pilot scale (Table 15). Corella, et al. studied calcined dolomite in the primary bed and nickel catalysts in secondary bed. They found the in-bed use of dolomite significantly changed the product distribution at the gasifier exit [47]. The dolomite was used in the secondary bed reformer where it performed slightly better in terms of tar reduction [148]. Although, dolomite has a significant activity for tar reforming, challenges such as deactivation and syngas quality prevent it is used alone as a catalyst. Nickel-based catalysts in the secondary bed reactor can efficiently remove tar to the required level. Nickel-based catalysts can only deal with the syngas with tar concentration less than 2.0 g/NM^3 and need a primary guard bed [149,150]. Novel catalysts demonstrate convincing total carbon conversion in biomass gasification [151,152]. Rh/CeO_2 catalyst exhibited far better performance than the others tested, however, the catalyst is deactivated due to the sintering [153]. Using SiO_2 as second support inhibits the agglomeration; but, it increases the cost of this intrinsically expensive catalyst. One low-cost char-supported iron catalyst was reported by Asadullah and Curtin. The catalyst applied in an especially designed gasifier showed much better performance in terms of tar reduction to less than 100mg/Nm^3 and burnable gas composition higher than 80 vol% [154,155,58,156].

Gasifier Type	Feeding rate (kg/h)	Catalyst type	Gas Composition (vol%)					Tar (mg/Nm^3)	LHV (MJ/Nm^3)
			H_2	CO	CH_4	CO_2	N_2		
Fluidized bed for wood [146]	20	Sand Primary bed	21.9	31.7	8.6	30.7	31.5	10.1	
Fluidized bed for wood [145]	20	Olivine$(MgFe)_2SiO_4$ Primary bed	19.4	30.1	8.9	36	53.1	12.8	
Fluidized bed for wood [146]	20	Magnesite($MgCO_3$) Primary bed	35.9	12.7	5.5	42.6	49	2.2	
Fluidized for Pinewood chip [47]	10	Calcined dolomite in bed	43	27	4.8	20		2000-3000	12.3
Bubbling fluidized bed for Pine wood chip [148]	10	Secondary bed dolomite	38	36.9	7.2	33		1720	15
Bubbling fluidized bed for Pine wood chip [150]	20-May	Commercial Ni catalyst	51-59	24-32	0.2-1.6	23-Sep		20-May	12-Oct
Up-draft for olive husk [157]	50 kg/h	Al_2O_3	14.1	13.9	2.9	13.2	55.9	98	

Table 15: Details of catalytic cracking data derived from different pilot-scale gasifiers [58].

Although the catalysts have advantages, such as high efficiency, environmental friendliness (no secondary pollution), low cost, and easy renewability, still research in the following areas worth to be done: (1) Energetically development of nano-catalysts because of their high tar reforming efficiency and excellent catalysis property; (2) Select reasonable catalysts according to the type of tar and its concentration; (3) Set up the recycling systems like chemical looping reforming (CLR) for tar elimination and catalysts regeneration; (4) Combination of catalytic tar elimination under higher temperature and tar adsorption with absorbents under ambient temperature [8].

Gas Cleaning Technologies-Economical View, Technical Barriers, and Future Opportunities

Syngas from biomass gasification can be potentially used for different applications: compressors, IC engines, gas turbines, fuel cells and liquid hydrocarbon fuels. For each application, the syngas needs to meet specific gas quality requirements. Biomass gasifiers have the potential to provide electricity in rural regions especially in the third world countries where biomass can be supplied from abundant agricultural industries and does not have access to the grid i.e. bubbling fluidized bed gasifiers (BFBG) has the potential to supply 100kWe energy that is enough to supply electricity for 100 households [4]. In waste-to-energy cogeneration plants, the cost-effectiveness of the plants depends on parameters such as plant capacity, a charge levied upon a waste provided (the gate fee), waste calorific value, including the percentage of moisture and biodegradable matter, syngas cleaning cost, hazardous waste disposal cost, revenue from the selling of electricity and heat, and loan condition. Ash and residue disposal could increase the cost considerably [158]. The biggest challenge in the commercialization of large-scale biomass gasification is the lack of cost-effectiveness and efficient remediation technology that removes the contaminants are formed during the gasification [58] include particulates (ash, char, condensing compounds), alkali metals (especially sodium and potassium in vapor phase), fuel-bound nitrogen (forming NO_x during combustion), sulfur, chlorine, and tar [4]. Producer gas cleaning system is very expensive and can cost up to half of the total system capital cost [85].

The syngas cleanliness depends on the required specification of different applications. However, no single clean method succeeds easily to meet the application requirement. Although many reports have provided information about the configuration and performance of the clean system for biomass-derived syngas cleaning; only a few literature studies have reported tar concentration in the cleaned syngas [75,90,159,160]. Hence, it's unclear whether the final tar content in the syngas is reduced to acceptable ranges or not. Usually, the cold gas cleaning process includes a combination of physical-mechanical gas purification systems. A combination of oil scrubber and char-bed filter can eliminate tar up to 98% [161], however, pressure drop should be considered on a larger scale. Unyaphan, et al designed a cleaning system that include cyclone, ceramic filter, air cooler, water coolers, venturi scrubber, and packed bed adsorber in series. Injection of produced gas in the form of microbubbles inside the venture oil scrubber increased the tar removal efficiency up to 97.7% [162]. Quench coupled with Adsorption Technology (QCAT) filling with ceramic rings and adsorbents with a mesh size of less than 24 were able to remove tar up to 99.9% and reach the tar concentration to less than 20 mg/m^3. Absorber regeneration could be a challenge [163]. Mixing wet vegetable oil scrubber as a physical removal method and dolomite guard bed as chemical tar removal technology reduced the tar 97% and reduced its dew point to 17° C. Pallozzi, et al. reported vegetable oil scrubber saturation time is very low and ~ 1.5h. The saturation time for dolomite guard bed is more than 14h but it is not able to reduce the dew point to less than 56° C. Mixing these two technologies can improve both saturation time to more than 12h and tar removal efficiency [164]. Up to date, the syngas clean technologies have been significantly developed for qualified gas production; however, commercialization of biomass energy is still challenging, mainly because of the tar issue. It needs a synergistic combination of developed efficient technologies for removing tar and other toxic gases and particles. Hence, syngas clean processing design and system configuration/optimization play an important role in syngas usage. The most concerns in syngas cleaning systems towards commercialization are total energy efficiency, cost, and environmental impact and the important parameters include gas composition, lower heating value (LHV), cold gas efficiency, waste product treatment, health hazard, and danger of fire and explosion, etc [67].

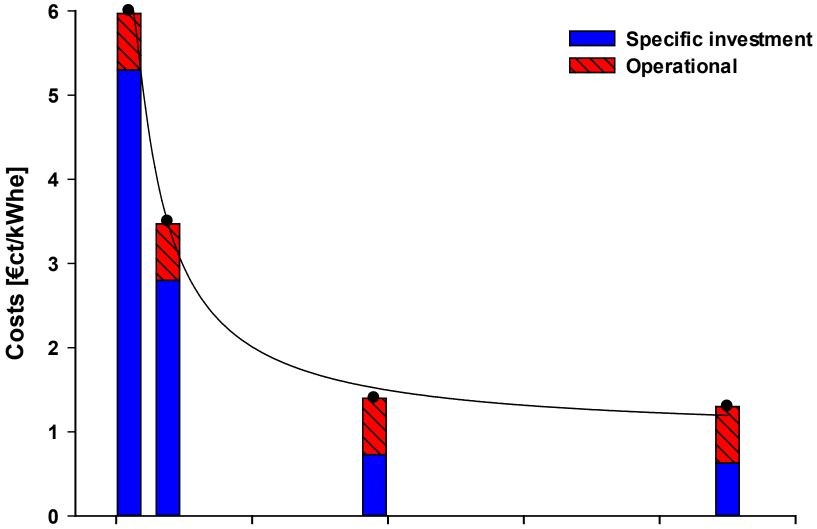
Primary treatments suffer from system complexity, limitation in the flexibility of feedstock and scale-up, compromised heating value, plugging of feeding, the production of waste streams, and a narrow operating window [20,165]. Hence, the suitable scenario for real-life experience is to reduce tar to a certain level by thermal cracking or catalytic cracking and simplify the system design, rather than to provide for very low tar emissions solely through primary treatments. In the gasifier, catalytic cracking is more advantageous in terms of energy efficiency compared to thermal cracking.

Among chemical and physical methods in secondary treatment to remove contaminants especially tar, chemical conversion methods including catalytic cracking, thermal cracking, and plasma gasification are more reliable and have higher efficiency but dry and wet mechanical/ physical gas cleaning methods are simpler and less expensive. Cyclone, rotating particle separators (RPS), electrostatic precipitators (ESP), bag filters, baffle filters, ceramic filters, fabric/tube filters, sand bed filters, absorbers, etc. are considered as dry cleaning methods and spray towers, packed column scrubbers (wash tower), impingement scrubbers, venturi scrubbers, wet electrostatic precipitators, OLGA, wet cyclones, etc. constitute wet cleaning methods [59].

Physical methods of secondary treatments produce disposals that reduce energy conversion efficiency [4]. Technical barriers for dry gas cleaning include the higher cost of temperature tolerable materials, tar removal efficiency, and plugging issues. Low-cost catalytic filters applied towards processing at a large scale are a promising direction for further technical development. Scrubbing with an absorbent liquid is a well-known technique for treating low-concentration, high-volume gas streams [4]. Wet gas cleaning processes remove various contaminants that are soluble in the chosen solvent. Different solvents such as water, oil, diesel fuel, engine oil, or fatty acid methyl esters derived from vegetable oil are used. Solvent efficiency, solvent price, effluent treatment cost, and process complexity are barriers in gas cleaning. Water as a cheap solvent is able to remove NH_3, HCl, H_2S, and light and oxygenated tar compounds but has low efficiency in removing non-polar heavy and heterocyclic tar [126]. Conventional wet scrubbers using water have disadvantages such as clogging, salt formation inside and outside of the nozzle, soaping, fouling, expensive sludge disposal and water treatment, and reducing the heating value of the gas [4]. Hence, although the tar removal efficiency of water scrubbers is acceptable, it is not a recommended technology for tar removal in the future. Effluent solvent treatment in a cold gas system causes the system to be more amenable to large scale applications in terms of cost, especially wastewater treatment [58]. Conventional wet scrubbers use water as a scrubbing medium; although, they are cheap they also consume a high amount of water. Providing cool water especially in summer and the necessary wastewater treatment make this technology expensive and complex. Water is only able to remove hydrophilic components (~ 30% of tar); so non-polar components that constitute the main part of the tar still remain in the syngas and cause clogging and fouling [4]. Organic solvents such as washing reagents that have high efficiency (>60%) in removing non-polar compounds are more expensive compared to water. Their tar removal efficiency depends on the kind of solvent, the initial tar concentration, and residence time. Organic solvents remove light tars that reduce the heating value of syngas. Removing light tars from the scrubber media by stripping and recycling them to the syngas can improve the heating value. The net energy efficiency of a wet scrubber is low. In a wet scrubber, salt forms in the tips, nozzle, tubes, and walls of a scrubber. Currently, OLGA is the only cold gas cleaning system that is claimed to be economically promising, highly efficient, without fouling and tar-free technology; however, the feasibility, durability, and reliability for the large-scale process with low cost are still under investigation. OLGA by removing tar without water condensation and recycling heavy and light tar consequently recovering all the energy and omitting tar waste stream minimized the water treatment cost and reduced the operating cost [166,85]. OLGA removes 99% of heavy and light tar. Cooling down the RME scrubber and OLGA high production cost may affect the cost feasibility of OLGA at a lower scale [73]. On a scale higher than 2000 m_n^3/h, the total specific costs are reduced to ~1.5 €ct/kWh_e. 99% of phenol and 97% heterocyclic tar are removed from wastewater by stripping [166]. So, there is no need to treat wastewater further [4]. OLGA technology is favorable at scales higher than 4000 m_n^3/h (corresponding to 10 MW biomass input); Boerrigter, et al demonstrated investment and operational cost at various scales (Figure 8) [166]. OLGA was originally built to remove tar from high-temperature gasifier (> 800°C). Condenser and absorber work with oil and water, respectively. At gasification temperatures less than 750°C, oil and water are replaced with two different oils to avoid tar condensation and polymerization, keep oil viscosity in a certain level, and guarantee polar tar removal [167]. A cyclone and a filter are used downstream to remove the tar [74]. Table 6 demonstrates some large-scale systems with the methods that have been used for water treatment and water recycling and Figure 8 demonstrates total investment and running cost to upgrade produced gas.

Catalytic filters are another novel mechanical treatment method used to minimize the cost of wastewater treatment due to the pollution by tar compounds [4]. Thermal and catalytic conversion methods completely destroy the tar; so, they have higher energy conversion efficiency. Also, they reduce waste streams and convert contaminants to environmentally benign or useful products [58]. Thermal cracking needs a supply of energy; to make it economical, the reaction conditions should be optimized [4,58]. Thermal tar cracking imposes a heat penalty [74].

In terms of thermal cracking methods, in order to improve the tar removal efficiency, theoretically, the effort can be made by increasing residence time, increasing temperature or promoting partial oxidation by adding air, or oxygen. However, it will largely increase the system and operational cost and decrease energy efficiency. Hence, the thermal cracking method is not the best choice for tar removal. The system design by applying thermal cracking methods should be based on the different gasifiers and take advantage of the high temperature of the gasifier or exit gas. If partial oxidation by adding air or oxygen is used, the balance between energy expense and whole process cost need to be considered.



M_n^3/h wet gas	190	760	3800		9000
MWth biomass	0.5	2	10		25
Total specific cost [€ct/kWhe]	6	3.5	1.4		1.3

Operational costs= 0.67€ct/kWhe (0.07 €ct/kWhe (utility) plus 0.6 €ct/kWhe (scrubbing liquid consumption))

Figure 8: OLGA system- investment costs vs. produced gas [78].

From an economic and technical point of view, the catalytic process is a promising alternative. The great advantage of this strategy is that a high degree of purity can be achieved at a lower temperature compared to thermal cracking. In addition, it has the potential to increase energy conversion efficiencies while simultaneously eliminating the need for the selection and disposal of tars [5]. Using a catalyst reduces the required energy, increases the tar conversion efficiency, and eliminates the need to collect and dispose of the tar [130]. Its major limitation is fast catalyst deactivation, catalyst disposal, and catalyst regeneration cost [58]. Ideally choosing a cheaper catalyst with high activity, stability at high temperature, high level of poisons acceptance, and less coke formation can improve syngas quality and lower system and operational cost [168]. Up to date, impregnation of natural catalysts such as olivine, dolomite, and zeolite with nickel provide a relatively inexpensive catalyst with higher stability and less coke formation [4]. Char-supported metal catalysts are also an encouraging way to achieve the goal of low cost and high efficiency [20]. Co-impregnation of nickel on natural catalysts (olivine, dolomite, and zeolite) provides a relatively inexpensive catalyst with reduced coke deposition on the catalyst and increased stability. Dolomites are the popular cheap catalysts for tar elimination. But they are very fragile and quickly eroded in fluidized beds with high turbulence. Olivine, similar to dolomite is cheap but has a higher attrition resistance. Its mechanical strength is comparable to sand, even at high temperatures. The catalytic activity of olivine is less than dolomite [169]. Other kinds of catalysts still need modification to be used in the catalytic cracking system. Non-nickel metal catalysts especially rhodium-based catalysts have high tar conversion efficiency but they are more expensive than nickel catalysts. Extending their lifetime is very necessary to make them more commercially viable. Acid and base catalysts produce lots of ash and the formation of coke deactivates the catalysts fast. Char and activated carbon are produced inside the gasifier and are cheap; but, coke blocks their pore too [4]. However, char could be replaced by the newly generated char in the same system regularly. Catalytic tar cracking is suitable for large-scale systems with high-temperature gasifiers and high tar levels [72]. In both thermal and catalytic cracking methods, waste stream can be an environmental barrier, and energy consumption affects the cost of the system [20].

Vecchione et al. compared a catalytic tar reforming system using a Nickel-based catalyst with a scrubbing system using oil and water as the scrubber media. Catalytic tar removal showed reliable performance (> 90%) at 800° C. However, catalyst deactivation due to sulfur, chlorine and alkali metals, coke formation and catalyst sintering due to repeated regeneration cycles at a high-temperature increase the catalyst consumption and disposal cost. Using water as a scrubber media brings down the purification treatment cost but the used oil in the scrubber can be used as a fuel in the same gasifier or transferred to another system and reduce the disposal and treatment cost considerably [85] (Figure 9).

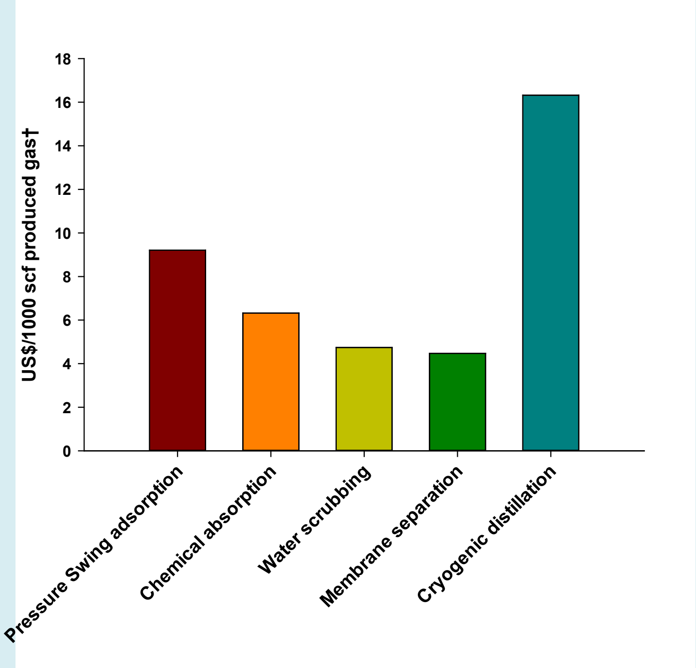


Figure 9: Total Investment and Running Cost to Upgrade Produced gas [170].
†Data for 130 – 161 Nm^3/h producer gas output rate

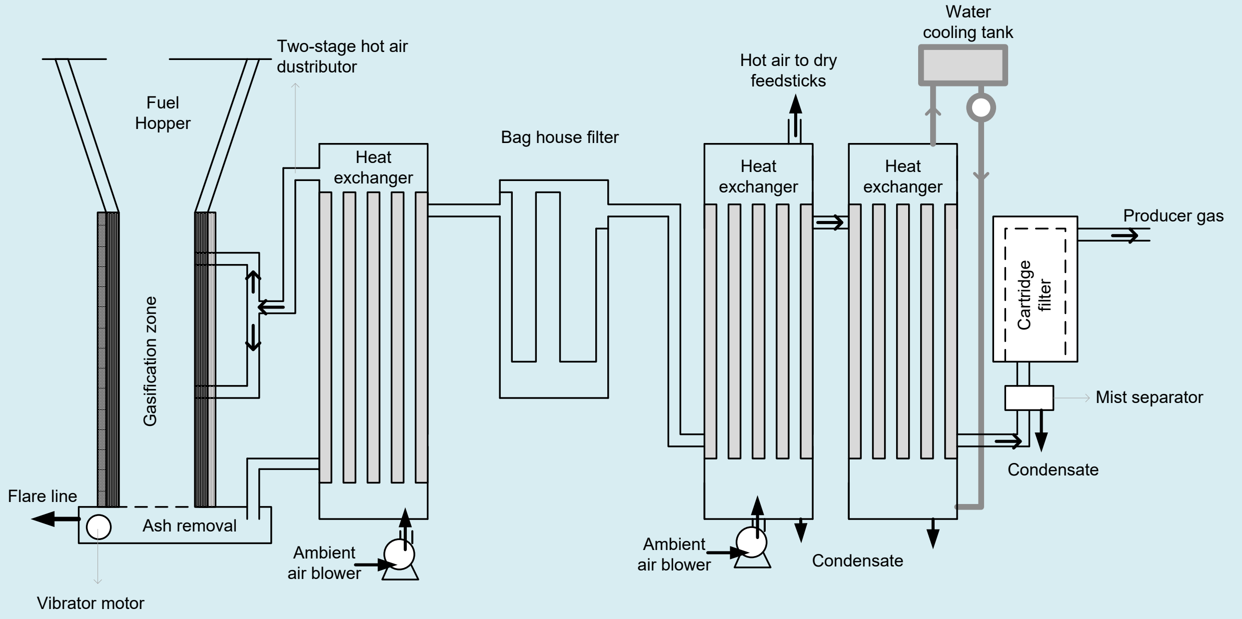


Figure 10: Dual fired gasification system scheme, adapted from [88].

Raman, et al. [88] designed a dual-fired downdraft gasifier to reduce the tar content of the produced syngas. The dual system helps to produce low tar syngas, with tar level reaching less than 100 mg/Nm^3. Moreover, producing the low tar content syngas eliminated the wet scrubber cleaning system and reduced the maintenance cycle 5-8 times. To remove the tar, the system was equipped with dry gas cleaning (filter) and indirect gas cooling (heat exchanger) (Figure 8). In this system, in the first stage, 70% of the required oxygen enters the reactor and produced moisture content charcoal, volatile matter, fixed carbon, and ash. In the second stage, 30% of the air is introduced to the bed vertically to provide the constant temperature. Air at high temperature inters this stage and its temperature was between 935° C and 1100° C that burn gas partially and cracks tar at high charcoal bed temperature. Using a dry gas cleaning system eliminates a large quantity of water usage and its disposal (Figure 10).

The Güssing system was developed for a small town in eastern Austria. Although the biomass is provided through the regional forest and provides energy for this town, still it was very expensive to use a gasifier as a source of energy. The system needs to be improved to become more reliable economically. In the Güssing method, biomass gasifies in a fluidized-bed steam gasifier at 850°C. A water heat exchanger reduces the temperature from 600°C to 150°C. Then the syngas passes through a fabric filter to remove dust. The removed dust is then recycled to the combustion zone. The downstream scrubber, which used biodiesel as a scrubber media, reduces the concentrations of tar, ammonia, and acidic gas components. The gas temperature reached 40°C after passing through the scrubber which is suitable for gas engine application. Part of the scrubbing agent is replaced with the fresh one continuously and the removed media is introduced to the combustion zone. The condensate is used to produce the steam needed in the gasifier. Accordingly, no residue or wastewater remains apart. The pre-coat material removed the chlorine. The system produces 2MW electricity and 4.5MW heat per year and its efficiency is 80%. Table 16 demonstrates the costs [68,171,172].

Particulars	Investment cost, €M	Funding (EU, national) , €M	Operation cost/ year	Heat, €-cents/kWhth		Electricity, €-cents/ kWhth
				into grid	Consumer	
Cost	10	6	10 to 15% of capital costs	2	3.9	16

Table 16: Güssing biomass CHP plant cost [173].

The Wiener Neustadt shaft gasification plant uses sedimentation and filtration to clean water or scrubber media. They recycle the sedimentation and filtration residues to the combustion zone [76,173]. The TU Graz system gasifies biomass in a double-fire fixed bed gasifier. In downstream a cyclone separates coarse particles. Then, the syngas is cooled down to 180°C in a heat exchanger to pass through a fabric filter for further particle removal. A scrubber which is operated with a mixture of water and RME removes tar. A thermal conditioning unit with stripping/desorption and adsorption treats the accumulated wastewater. The treated water is subsequently pumped into the sewer system [68]. Pyroforce fixed-bed gasifier plant stores the scrubbing media temporarily in a condensate tank and is then disposed of. DTU (Danish Technical University) Graz gasifies biomass in two stages. First, the biomass is pyrolyzed, and then the produced tar and volatiles oxidize partially. The tar concentration in the syngas is as low as 25 mg/Nm^3.

Producing syngas with less tar (primary method) and using chemical and physical methods (secondary method) to remove tar are both required to reduce provided syngas for further application. One method cannot be efficient enough to reduce the tar to the required level. In both cold and hot gas cleanup, multi-contaminant removal would be desirable to drive costs down [58]. Accordingly, a mixture of various methods is required to reach the needed level of tar economically. Besides tar, other contaminants exist that need to be removed to some extent like particulates, soot, NOx, sulfur and chlorine, and alkali metals. In removing the alkali salts electrostatic and bag filters or wet scrubbers are very efficient [157]. Nitrogen components such as NH_3 and HCN that have a high solubility in water and are difficult to remove them are better to be removed before combustion or wet scrubbing. Standard catalytic methods for NOx treatment are able to remove nitrogen compounds easily [157]. In choosing a suitable technology, besides efficiency, the global cost plays an important role. Equipment investment costs decrease by increasing the produced gas scale; so, small-scale produced gas (0 – 100 Nm^3/h raw produced gas) is usually very expensive. Installing a large central processing facility brings up the piping cost and introduces the technical challenges with regards to raw produced gas transportation such as pipe corrosion. However, one or more bulk units that include the entire upgrading system that only requires piping and wiring connections are the best solution to make the system economically. Increasing energy efficiency and improved contaminant removal and resistance can improve the system. In addition, more research work needs to be done in the area of design and optimization of syngas cleaning systems. Gas cleaning conditions and systems configuration to control the tar levels need to be continuously modified for better efficiency and cost-effectiveness.

Conclusions

As a renewable energy source, biomass has the potential to displace petroleum and other fossil fuels via generating syngas for fuel, energy, and products through gasification. Although significant improvement has been made, it seems still challenging to achieve commercial deployment of biomass gasification at a large scale which is reliable and durable in real integrated gasification (IG) based system environment, because of the syngas contaminants. The importance of syngas cleaning, especially targeting tar removal has driven research that aims to develop gas cleanup strategies that will improve efficiency and decrease cost for many decades. Many technologies have been developed to remove virtually all contaminants down to desirable lower concentration limits. Primary treatment cannot be counted on solely for tar removal mainly because of system complexity and inconvenient features. Physical methods especially wet gas cleanup technologies are effective, but not beneficial to reduce the overall efficiency due to syngas cooling, cost efficiency, and environmental sustainability due to post-treatment of toxic effluent streams. Based on this review, catalytic cracking of secondary treatment is more advantageous in terms of energy efficiency compared to thermal cracking. The dominant challenge in hot gas cleanup at demonstration or pilot scale is rapid catalyst deactivation and coking formation. This challenge is persistent due to the lack of thoughtful consideration to deactivation in many catalytic hot gas cleanup studies and design/optimization of syngas cleaning systems.

Recommendations

1. It is recommended that serious attention be devoted to evaluating catalyst resistance to deactivation in hot gas cleanup and also to strategic system configurations.
2. Using hybrid method can provide a solution to reach the needed level of tar, economically.
3. Beside total energy efficiency, and cost, the environmental impact of the system should be considered.
4. On developing the tar removal method, there should be concentration on the methods that produce less/no waste.
5. Scale of the gasifier should be considered in choosing the gas clean-up method.
6. Removal of contaminants such as particulates, soot, NOx, sulfur and chlorine, and alkali metals should be studied.

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