

Review

Lignocellulosic Biomass Gasification: Perspectives, Challenges, and Methods for Tar Elimination

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Abstract: Gasification of lignocellulosic biomass has been widely highlighted as one of the most robust and promising low-carb approaches toward sustainable energy production. The gasification syngas obtained from agro-industrial residues can produce heat, power, biohydrogen, and other drop-in biofuels via F-T (Fischer-Tropsch) synthesis. However, the tar formation during the thermochemical process imposes severe limitations on the commercial scale of this technology. Tar elimination is a critical step for avoiding damage to equipment and not restricting the further application of syngas. In this context, this work sheds light on the biomass gasification field and reviews some aspects of tar formation and technologies for its reduction and removal. The approaches for dealing with tar are primary methods, which suppress or remove tar within the gasifier, and secondary methods, which remove tar in post-operation treatment. Catalytic reforming offers the most cost-effective pathway to removing tar. The bimetallic combination of nickel with other metals and using biochar as support have been intensely investigated, showing excellent tar conversion capacity. Recent research has provided new trends in non-thermal plasma-catalyzed biomass tar reforming. Future studies should focus on the integration of catalysts with multiple techniques to improve efficiency and reduce energy consumption.

Keywords: gasification; biomass; lignocellulosic residue; syngas



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1. Introduction

With the world facing unprecedented environmental crises, the expansion of clean energy to phase out fossil fuels is now unquestionably urgent, at scale and speed. Due to the massive volume of greenhouse gas emissions, burning fossil fuels is by far the most significant contributor to intensifying the problems of global warming and environmental damage [1]. Nevertheless, coal, oil, and natural gas are still the primary energy resources used to supply domestic and industrial demands for power and fuels [2].

Renewable energy capacity has been growing faster than ever. However, the sustainable transition is not happening quickly enough to keep pace with the decarbonized

scenario [3]. To slow down the effects of climate change, the green transition must happen in all sectors of supply chains, not only concentrated in electricity generation. Investing in technologies that, besides electricity, produce drop-in fuels and bioproducts is essential to decarbonizing hard-to-abate sectors. In that context, byproducts from lignocellulosic biomass thermochemical conversion can supply heavy industries (steel, cement, petrochemicals) and heavy-duty transports (trucking, shipping, and aviation) [4]. Using lignocellulosic materials is a highly strategic way to address environmental and economic issues because it involves the recovery of agricultural and forestry residues as bioenergy feedstock [5].

Among thermochemical conversion processes (combustion, pyrolysis, liquefaction, and gasification), gasification is a key technology for exploiting biomass potential. The process involves a complex set of reactions that occur at high temperatures and in the presence of an oxidizing agent. The lignocellulosic biomass structure (cellulose, hemicellulose, and lignin) is broken into a gaseous mixture of H_2 , CO , CO_2 , and CH_4 . Biomass gasification also involves the formation of undesired products. Solid residue char, nitrogen and sulfur compounds (NH_3 and HCN , H_2S , COS , and CS_2), and tar are impurities limiting biomass gasification expansion [6].

The gas mixture from gasification is called syngas when it is basically composed of CO and H_2 and named producer gas when it has significant quantities of CH_4 , CO_2 , N_2 , and impurities. Producer gas has limited applications and is normally destined to be used as a fuel gas for turbines and gas engines in heat and power applications. Syngas can be used directly as fuel for heating and power generation or as an intermediate for producing biofuels and chemicals, such as biohydrogen, ammonia, and methanol [7].

Tar is a major concern in syngas applications. Its concentration must be controlled to meet the different tolerance levels for syngas utilization. Tar impacts thermal systems, reducing some of the available effective energy and leading to severe operational issues. Soot formations, corrosion, fouling, and abrasion of pipes and downstream equipment are among the causes related to the tar presence [8]. Common gasification systems yield a formation of 1–150 g/ Nm^3 of tar. This concentration makes the gas unsuitable for use in turbines and internal combustion engines or in producing fuels and chemicals. The recommended tar tolerance for internal combustion engines is below 100 mg/ m^3 , and it is suggested to be less than 50 mg/ m^3 for gas engines [7]. Therefore, the removal or cracking of tar is essential before applying biomass-producer gas.

Considering the vast role that lignocellulosic biomass thermoconversion is set to have in the decarbonization scenario, several in-depth studies have been conducted to understand tar's formation and mechanism. Some strategies have been developed to promote the removal (secondary methods) [9,10] or suppression (primary methods) [11] of tar. The main purpose of this work is to contribute to developing these technologies by elucidating some of the major barriers limiting gasification scale-up while exploring the more recent tar treatment technologies. This study focuses on exploring the mechanism and main precursors of tar formation. The role of the catalytic approach is extensively discussed, and new insights into plasma technology for tar reforming are reported. The paper is organized as follows: Section 2 provides an overview of biomass and bioenergy; Section 3 reviews gasification principles and syngas applications; Section 4 describes tar properties, formation mechanics, and methods for its removal; and prospects and conclusions are in Section 5.

2. Biomass and Bioenergy

2.1. Lignocellulosic Biomass

Biomass has been recognized as a highly strategic resource for providing clean fuels, heat, electricity, and chemicals [12]. Biomass is a carbon-rich material that, unlike fossil

fuels, does not add carbon to the atmosphere. The gases released when biomass is burned were earlier captured during plants' photosynthesis. Bioenergy is also a renewable energy source that benefits the best from the existing infrastructure based on fossil fuels [13].

Lignocellulosic residues from forest and agricultural activities can yield readily available biomass for energy purposes. Tons of straw, stalks, leaves, husks, and other wastes are generated worldwide all year long [14]. Residual biomass is a key element in supporting socioeconomic development. It offers a low-cost energy source and does not compete directly with food and feed applications or other industrial production.

Modern uses and conversion of lignocellulosic residues are challenging because of its complex and integrated network of structural biopolymers. The potential conversion of biomass into energy, fuels, or chemicals, as well as how the conversion process should be conducted, is totally dependent on the raw biomass's lignocellulosic composition.

Lignocellulosic biomass has low reactivity and a highly heterogeneous composition that comprises different contents of cellulose, hemicellulose, lignin, extractives, lipids, proteins, sugar, amide, water, ash, and other components. Each molecule plays a specific role in the cell wall and occurs in different proportions according to the biomass [13].

Cellulose, the main component of the plant cell walls and the most abundant biopolymer on Earth, consists of an extensive linear chain with a high degree of polymerization and high molecular weight [15]. It comprises D-glucose monomers in β -1,4-glycoside linkage stabilized by inter- and intra-molecular hydrogen bonding. This configuration forms microfibrils, which aggregate to form fibrils that will form the so-called cellulose fibers [16]. Due to this network, cellulose has a semi-amorphous structure with a highly crystalline region, conferring resistance to hydrolysis (organic solvent and water) and traction [17]. The degree of polymerization and crystallite varies with biomass species and types (Figure 1).

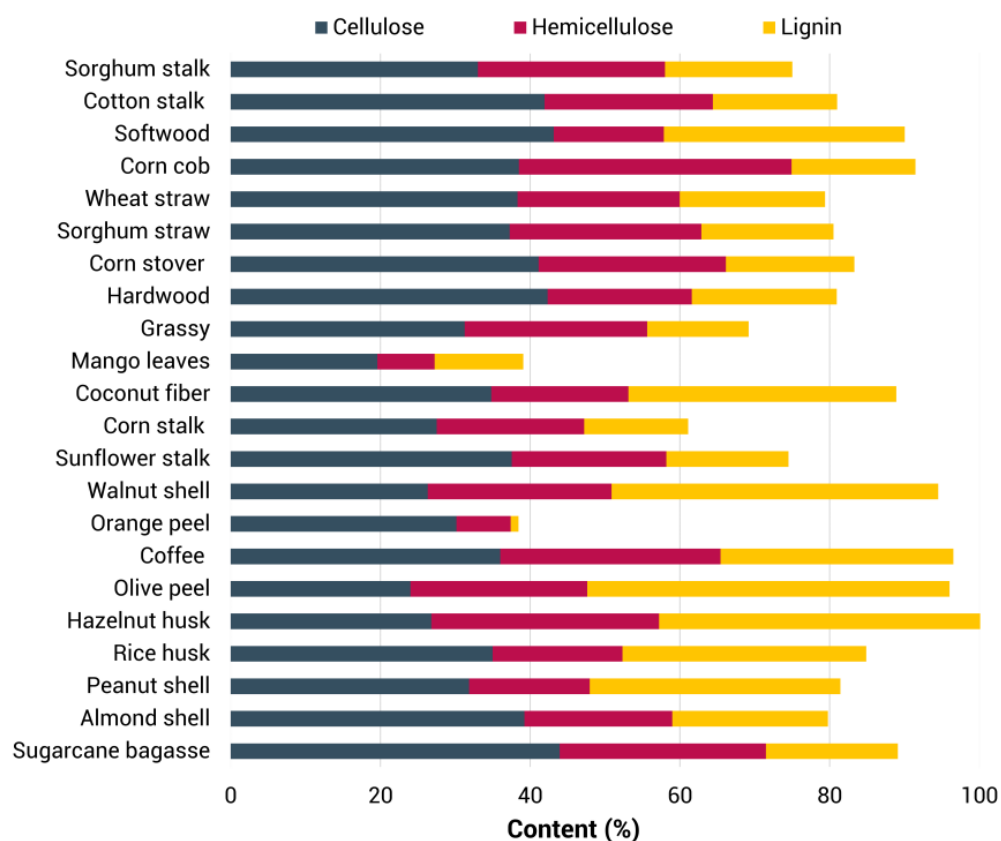


Figure 1. Lignocellulosic distribution in woody, grassy, and agro-industrial biomass. Data from Kammoun et al. [18], Lepage et al. [19], Paranhos et al. [20], and Raj et al. [21].

Hemicellulose is a carbohydrate polymer that, contrary to cellulose, has an amorphous, branched, and heterogeneous structure. It has a low molecular weight and degree of polymerization. Therefore, it is more chemically reactive and easier to hydrolyze [13]. Hemicellulose varies greatly with biomass sources since it can comprise different monomers such as xylose, arabinose, mannose, rhamnose, glucose, galactose, and uronic acids [22,23]. In general, softwood biomass is mainly comprised of hexoses (glucomannans and galactoglucomannans). They are more thermally stable and are degraded into formic acid, 5-hydroxy-methyl-furfural (HMF), and acetylfural [24,25].

The third molecule (lignin) is highly branched, the most complex and thermally stable of the three biopolymers. Lignin is a three-dimensional network resulting from the random polymerization of many cross-linked phenylpropane units. Its role in the cell wall is to protect and provide recalcitrance to the plant against physical and biological attacks and act as a cement by linking covalently to the hemicellulose to form a matrix that provides support to the cellulose fibrils [22,23]. The aromatic structure of native lignin is primarily made up of p-hydroxyphenyl (H), guaiacyl (G), and syringyl (S) monomers. However, the origin, type, or species of biomass influences the relative amount of each subunit. Softwood lignin is mainly G-type, and hardwood lignin contains a similar content of both S and G units. All three units are present in herbaceous lignin, but the H/S/G ratio also varies with biomass species and types [26].

Because of the differences in the chemistry and chain conformity, the lignocellulose building blocks differ in their function in the cell wall and change significantly in terms of thermal behavior and decomposition routes [27]. Due to its aromatic profile and various oxygen functional groups, lignin decomposes throughout a wider range interval (160 and 900 °C) with peak temperature above 400 °C [28]. The decomposition of hemicellulose (200–380 °C) and cellulose (320–400 °C) occurs in lower and narrower temperature ranges.

Yu et al. [29], Michailof et al. [30], and Chen et al. [31] demonstrated that hemicelluloses contribute significantly to the formation of gases and water vapor, while cellulose is preferred fragmented into levoglucosan and light oxygenates, important precursors of the liquid fraction (oil and tar). Lignin derivatives are found to influence the formation of solid and gas phases. Lignin was shown to be more likely to produce H₂, CH₄, and CO₂ (in gases) and generate liquids with the dominant presence of phenolics, alcohols, and aromatic hydrocarbons such as catechol, vanillin, methyl propanol, cresol, methoxyphenol, eugenol, methoxy propenyl phenol, and benzene [32].

2.2. Biomass Conversion Approaches

Due to biomass variability and versatility, as well as its potential for many usable forms of energy, several pathways can be applied to accomplish its conversion, i.e., physical, chemical, biochemical, and thermochemical [12,33].

All thermochemical processes involve heating biomass under controlled conditions. According to their associated oxidation environment, they can be broadly divided [34,35] to completely oxidize the fuel. Gasification involves using a sub-stoichiometric amount of oxygen for partial oxidation. Pyrolysis and direct liquefaction occur in the absence of oxygen.

Combustion is the most used conversion pathway in domestic and industrial scenarios because it is simple and well-established [36]. In combustion, reactions take place between the oxygen of air and biomass to release water, CO, and heat, which is used for heat and power. Combustion systems vary from small installations with primitive technology to modern facilities with highly controlled furnaces, viz. Combined Heat and Power (CHP) plants (efficiency of about 75%) [37]. Combustion projects have made important advances in their burning efficiency over the years; however, they are generally less effective than

other thermochemical technologies, besides contributing to local pollution by emitting CO₂, SO₂, and NO_x [38].

Pyrolysis is carried out under an inert atmosphere and at temperatures ranging between 400 and 600 °C. The process is conducted for the thermal cracking of biomass into liquid (bio-oil), noncondensable gases (biogas), and solid carbon-rich residues (biochar) [39]. Depending on the operating conditions, pyrolysis is categorized as slow, intermediate, fast, or flash, wherein the heating rate and residence time shift from 1–1000 °C/s and from seconds to hours, respectively [40]. The main product of pyrolysis, labeled bio-oil, consists of a mixture of oxygenated organic compounds from the decomposition of lignin (phenolics and hydrocarbons) and cellulose (aldehydes and furans) [31,41]. Applications on a commercial scale are still limited because of its high viscosity, acidity, and low stability. However, several scientific studies have focused on upgrading oil properties to be used as an alternative to petroleum derivatives [42,43] and act as an absorbent material to remove pollutants from effluents.

Hydrothermal liquefaction (HTL) thermoconversion occurs at temperatures below 400 °C, employing high pressures and using water (or organic acids) as solvents and hydrogen donors. Its mechanism and products are similar to pyrolysis. Biomass samples are fractionated into an oil phase, a water-soluble fraction, a residual solid, and gases [44]. The bio-crude oil has higher quality and energy density than pyrolysis oil. HTL oil does not present sugars and has minimal acids compared with pyrolysis oils. It has the potential for use as a transportation fuel after upgrading. Due to the liquid medium, HTL allows for the efficient use of biomass without prior drying. While pyrolysis is indicated for processing wet biomass, HTL is attractive for processing biomass with high moisture content, such as municipal and industrial stream wastes [45–47]. The elimination of the drying step also reduces energy consumption.

Pyrolysis and HTL have a huge role in decarbonization, producing valuable products from low-cost biomass. In any case, to meet a broader range of demands, syngas, a gasification product, is energetically more strategic. Bio-oil has a wide range of potential applications as fuel but requires upgrading for enhanced fuel properties and stability. Syngas holds significant potential as an energy carrier. It may require purification but can be used in power generation, chemical synthesis, and industrial heating. Synthetic natural gas and biohydrogen obtained from syngas are promising renewable, carbon-neutral alternatives to fossil fuels [48]. The gasification process and syngas will be examined in detail in the next section.

3. Thermochemical Conversion: Gasification

Gasification is a thermochemical technology that converts biomass into combustible gases under elevated temperatures and in the presence of sub-stoichiometric quantities of an oxidant agent, which can be air, oxygen, water vapor, or a mixture of them. The process can be divided into four basic stages: drying, which removes the surface moisture by vaporization (100–150 °C), pyrolysis, oxidation, and reduction (Figure 2) [49].

In the pyrolysis zone (200–700 °C), biomass initiates depolymerization via dehydration, decarboxylation, and decarbonylation reactions in the absence of oxygen. The process results in the release of permanent gases (CO, CO₂, H₂, and CH₄), condensable volatiles (naphtha, light hydrocarbons, and oxygenated compounds such as phenol and tar), biochar (nearly 10–25% of initial carbon content), and ashes [40]. Cellulose, hemicellulose, and lignin contribute in different manners to the formation of gases, liquids, and solids. Then, the properties and volume of each output product are mainly defined by the initial composition of biomass.

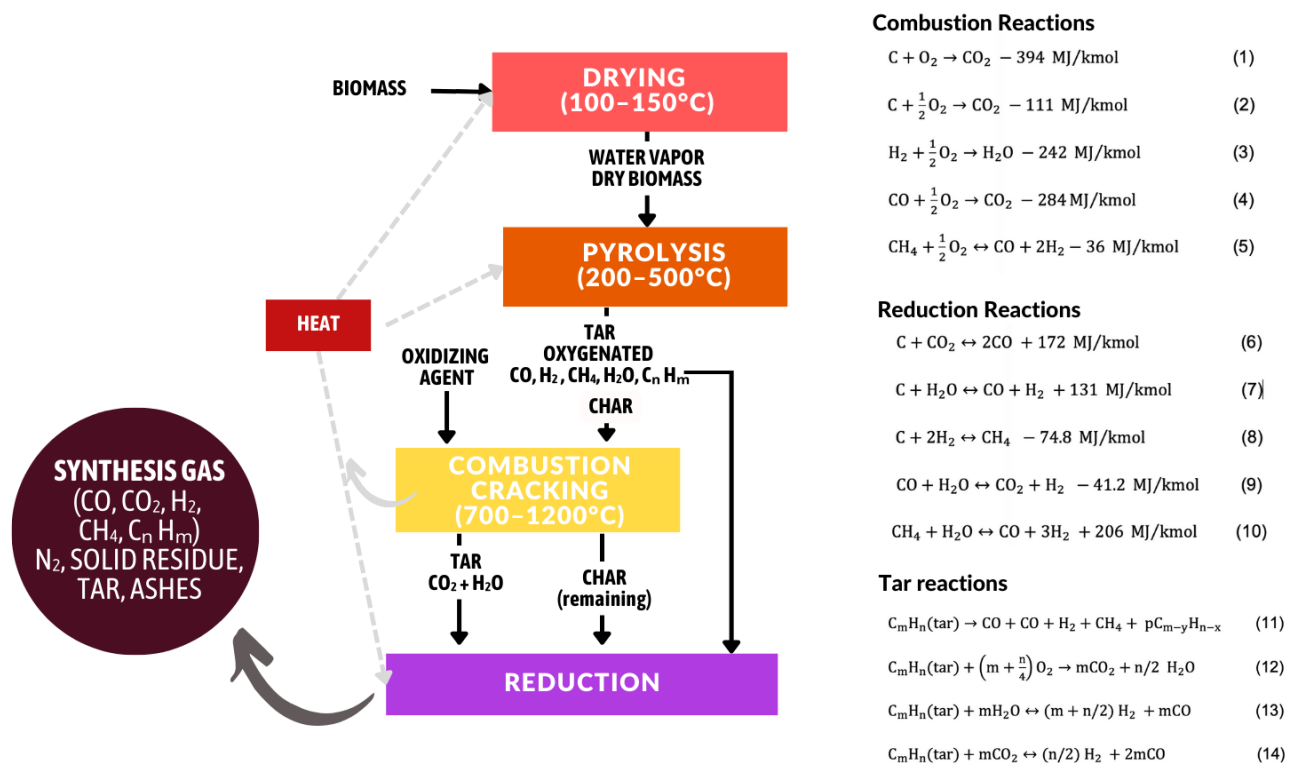


Figure 2. Distinct zones involved in biomass gasification with specific temperature ranges.

The combustion stage occurs in parallel and dynamically with the reduction zone. Combustion is the only endothermic zone of the whole process. It is in charge of providing heat (directly and indirectly) for drying, the linkage rupture during pyrolysis, and gasification reactions (Combustion Reaction, Figure 2) [50]. The reduction zone is characterized by an oxygen-poor environment. The reactions between the hot gas mixture and solid carbon, reactive and homogeneous reactions, are triggered to maximize the production of CO and H₂. The main reactions are Boudouard, water/gas reaction, hydrogenation, water/gas shift reaction, and steam/methane reforming (Reduction Reaction, Figure 2) [51].

When gasification is finished, the main product obtained is a gaseous mixture of H₂, CO, CO₂, CH₄, and other hydrocarbons. Some unwanted compounds, such as inert nitrogen, traces of H₂S, HCl, HCN, and NH₃, char condensable hydrocarbons (tar), and particulate matter, such as ashes and inorganics, are also present [52]. Tar is a subproduct of gasification formed in the initial stages when biomass undergoes pyrolysis. Reactions involved in tar formation and evolution include thermal cracking (>100 °C), oxidations, steam reforming, and dry reforming (Tar Reaction, Figure 2) [51,53].

Producer gas containing substantial amounts [54,55] is of poor quality and has limited uses for heat and power applications.

Syngas can be used as a fuel gas and as an intermediate product for the synthesis of more complex chemical compounds, liquid fuels, and hydrogen (Figure 3) [56]. However, each of these applications has tolerance levels of impurities and requires specific ratios among main gases. Therefore, the output gas often needs to be purified to meet the quality criteria. The severity of the cleaning process and gas treatment are defined based on both the final destinations' requirements and the quality of the raw gas that leaves the reactors, whether in the matter of gas ratios or the presence of impurities.

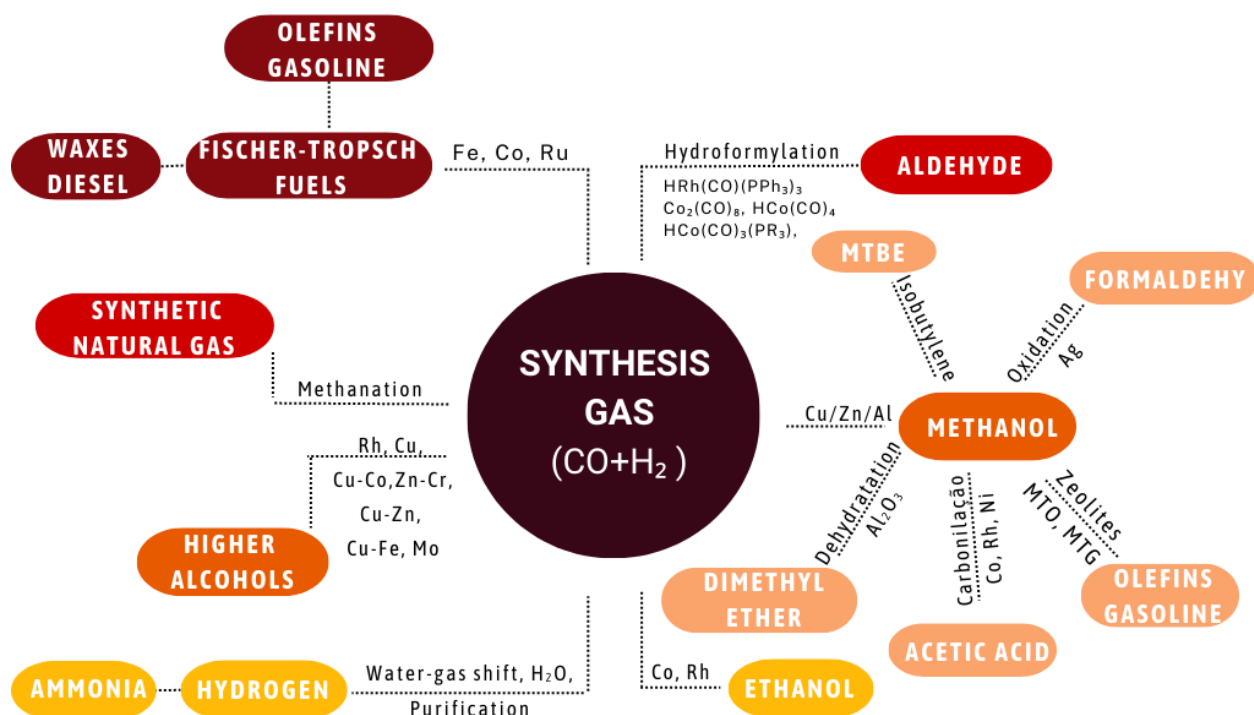


Figure 3. Synthesis gas destination. Source: authors based on Sikarwar et al. [57].

Among all the unwanted compounds, tar is the most difficult to remove from syngas (Table 1). Tar is known for its complex nature (of aromatic compounds such as benzene, naphthalene, toluene, anthracene, and pyrene) and tendency to undergo polymerization [58]. All parameters of the thermoconversion process influence the final tar concentration and composition. Because of the problems with tar presence, researchers have attempted to study the effect and how to adjust the various process variables to quantify and control tar formation. Other studies have been dedicated to developing new methods to clean up gas and deal with tar present in gas streams.

Table 1. Desirable syngas characteristics according to destination.

Destination ¹	GT	FT	Methanol	IC	BH ₂
Compounds					
Tar (mg/Nm ³)	<0.5	<0.1	<1	<100	Low
Sulfur (ppmv)	<20	0.01	<1 ⁴		Low
Nitrogen (ppmv)	<50	0.02	0.1 ⁴		Low
Alkali (ppmv)	<0.02	0.01	<10 ⁵		Low
Halogens (ppmv)	<1	0.01	0.1		Low
Particulate ⁶ (mg/Nm ³)	<30	<0.02	<0.02	<50	Low
H ₂ /CO	NR ²	2	2	NR	High
CO ₂	NC ³	Low	Low	NC	NR
N ₂	NC	Low	Low	NC	Low
CH ₄	High	Low	Low	High	Low

Source: Ciferio and Marano [59], Rios et al. [60], and Woolcock and Brown [61]. ¹ GT—Gas Turbine; FT—Fischer-Tropsch; IC—Internal combustion; BH₂—biohydrogen. ² Not relevant; ³ Not critical; ⁴ mg/Nm³; ⁵ ppb; ⁶ ashes, char, and solid residues.

4. Tar Chemistry and Removal Methods

4.1. Tar Classification

Tar is a result of several heterogeneous reactions, presenting complex and non-predictable compositions. It is commonly classified into four groups according to the

process conditions in which compounds are formed: primary tar, secondary tar, alkyl-tertiary, and condensed tertiary tar [62].

Primary tar comprises oxygenated compounds released from cellulose, hemicellulose, and lignin devolatilization followed by cracking reactions. As described in Section 3, during gasification, at 400–600 °C, biomass macromolecules start depolymerization into oxygenated and condensable organic compounds containing acids, aldehydes, carbonyls, alcohols, and ketone functional groups [63]. Primary tar from lignin mainly contains phenolic fragments from the units present in the original aromatic structure, e.g., guaiacol and syringol. Holocellulose contributes to anhydrosugars [64,65].

Primary tar is refractory to thermal and oxidative cracking reactions [62]. Then, when temperatures increase, secondary tar reactions (dehydroxylation, demethoxylation, and demethylation) take place to generate secondary and further tertiary tar. This results in the formation of heavier molecules and a more condensate profile [63].

The secondary tar is formed by cracking reactions of lignocellulose derivatives at 600–850 °C. The oxygen-free components became more abundant, generating more aromatic skeletons. It comprises branched and heteroatom compounds—heavier single-ring aromatic molecules such as benzene, cresols, toluene, xylene, and styrene [66]. Reaching temperatures higher than 800 °C, primary and secondary tars are rearranged by ring condensations and polymerization into tertiary tar (generally formed once primary tar is fully decomposed).

Between 850 and 900 °C, peak production of tertiary-alkyl (e.g., 2-methyl-naphthalene, biphenyl, and 2-ethenyl-naphthalene) occurs. However, the predominant trend is condensed aromatics evolution for stable polycyclic aromatic hydrocarbons (PAH) compounds such as naphthalene, anthracene, phenanthrene, and pyrene [65]. Tertiary-PAH tar has high thermal stability and is the most difficult to eliminate, and is a precursor for soot formation. The technical difficulties in scaling up gasification are attributed to this tar class [67]. Suppressing the formation of tar precursors and controlling the development of PAH tar can be said to be a critical step in promoting the efficient utilization of biomass.

In addition to primary, secondary, and tertiary categories, more specific compound classifications have been considered for tar. This is because some organic acids, classified as primary products, may present a different behavior and persist longer than other primary products [62]. A classification based on the size and number of aromatic ring structures is provided by the Energy Research Centre of The Netherlands (ECN). Tar is grouped into five classes: class 1—GC undetectable (very heavy, seven or higher ring compounds); class 2—heterocyclic (phenol, cresol, and pyridine); class 3—light aromatics (toluene, styrene, and xylene); class 4—light polyaromatic (naphthalene, phenanthrene, and anthracene); and class 5—heavy polyaromatic (fluoranthene, pyrene, chrysene, perylene, and benzo(ghi)perylene) [68].

4.2. Tar Formation and Evolution from Lignocellulosic Biomass

The detailed formation mechanism and final composition of tar are largely unclear. However, some proposed mechanisms and identified patterns are reported in the literature (Figure 4) [69,70]. Tar derived from holocellulose contains significant concentrations of light tar, such as furfural, furfural alcohol, acetoxyacetone, cyclopentenone, and cyclopentadiene. Otherwise, benzene and tertiary-PAH tar formation is influenced mainly by the presence of lignin. Some celluloses can also contribute to releasing aromatic precursors and high-activity small-molecular gases (ethylene and acetylene) [71]. Due to the aromatic structure, lignin is often the most decisive component that controls tar formation and the rate of production [65].

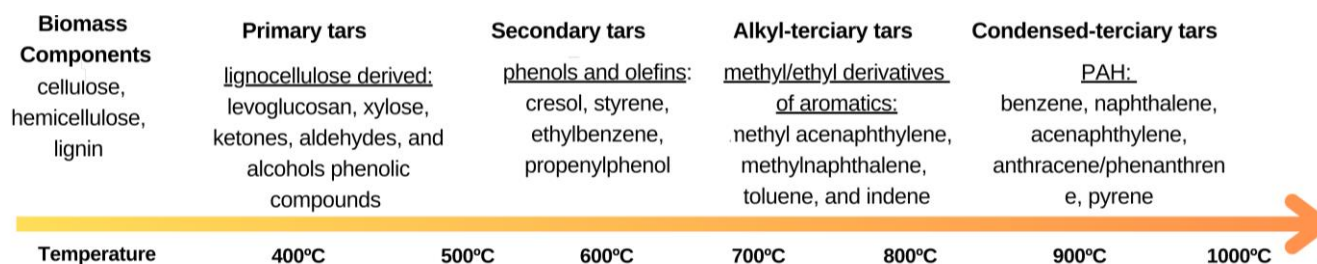


Figure 4. Tar formation and evolution from lignocellulosic biomass components.

Anna Trubetskaya et al. [52] studied the fast pyrolysis of cellulose, hemicellulose, and two different lignins. The authors found lignin to be the primary contributor to tar yield: up to 80 mg of tar per g of the sample compared to a maximum of 40 mg/g for holocellulose. They also identified lignin tar as PAHs, methyl/ethyl phenols, and methyl/ethyl PAHs. Meanwhile, long-chain acids, paraffins/olefins, and miscellaneous hydrocarbon compounds (HCs) were considered as derived from the holocellulose.

The two main pathways to form PAHs are (i) through the direct combination of one-ring species benzene and phenol and (ii) by acetylene addition followed by cyclization at pyrolysis conditions. Benzene formation is a result of reactions involving C_2H_2 , C_3H_3 , and C_4H_6 , in which H, OH, CH_3 , and C_6H_5 are key radicals [72]. PAHs from benzene are related to the amount of lignin. They are formed by ring condensation, hydrogen abstraction, and acetylene addition. YH Qin et al. [73] state that the holocellulose content strongly correlated to the phenol pathway due to the oxygen-containing compounds and unsaturated alkenes. During gasification, phenol mostly undergoes decomposition with O-H bond dissociation, followed by ring-opening to produce smaller gas products. However, some phenol may also be converted to naphthalene by decarbonylation followed by a Diels–Alder reaction [74,75]. In the phenol pathway, CO radicals of phenolic compounds are lost to form cyclopentadiene that generates cyclopentadienyl. Further, they are combined by radical self-reaction into aromatic compounds from naphthalene to chrysene [76–78].

4.3. Tar Destruction and Removal

Several approaches have been developed and employed to deal with the tar formed during gasification. The methods are generally classified into primary (in situ) and secondary (ex situ) methods (Figure 5). Primary methods occur inside the reactor itself during operation. It is based on (i) the control of tar formation by finding optimal operation conditions and improving reactor design or (ii) the use of catalysis to improve tar cracking/reforming to light hydrocarbons and permanent gases, mainly H_2 and CO. Secondary methods occur outside the reactor. Tar is removed using physical/mechanical separation techniques or by destruction in catalytic or non-catalytic thermal cracking and partial oxidation downstream of the gasifier.

Through in situ techniques, the producer gas is expected to meet quality standards without further tar treatment, reducing operation costs. However, in some cases, the combination of one or more in situ and ex situ may be necessary to remove all tar [79]. The methods and strategies developed to deal with tar will be described according to operational conditions, gasifier design, catalyst, and physical methods. Table 2 summarizes tar efficiency removal using different approaches.

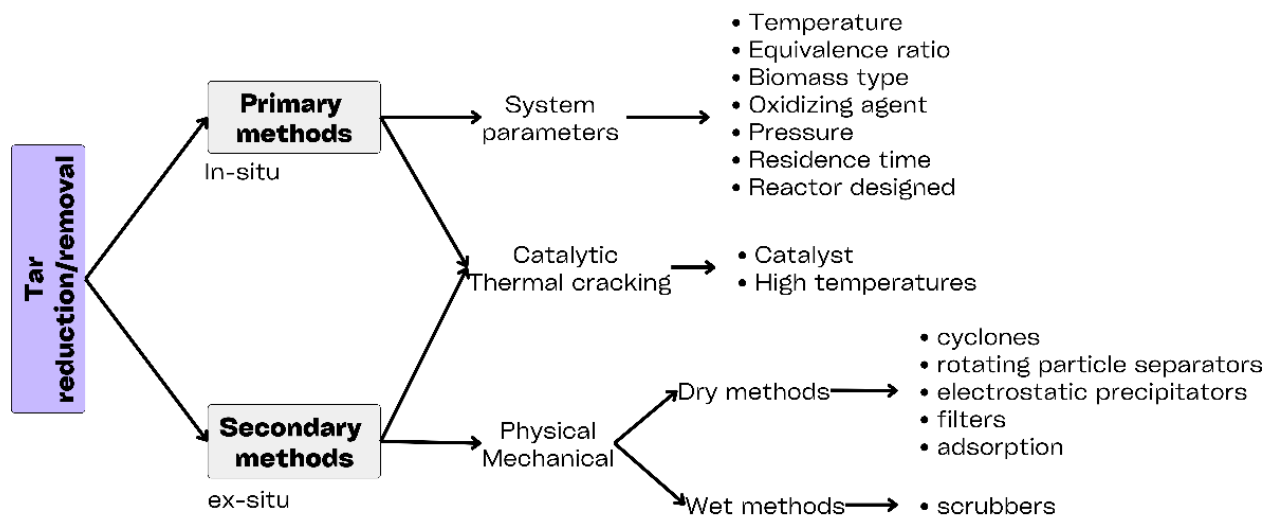


Figure 5. Tar removal methods.

Table 2. Final tar content and abatement efficiency based on different cleaning or removal methods.

Sample	Process	Cleaning/Removal Method	Tar (g/Nm ³) BC/AF Efficiency	Ref
Spruce wood	Air gasification Downdraft reactor T: 850 °C	Cyclone and packed bed filled with pine chips	1.6–1.4 12.5%	[80]
Pine 8% moisture		Cyclone and packed bed filled with pine chips	3.9–2.5 35%	
Pine chips		Wet packed bed scrubber using chips as the packing material and waste cooking oil as the media	1.4–0.3 78.5%	
Palm kernel shell	Steam gasification T 750 °C S/B: 2.25	In situ catalyst zeolite	0.7 98%	[81]
	T: 850 °C S/B: 2.25		0.12	
	T: 750 °C S/B: 1.5	In situ catalyst zeolite acid leaching	4.6–1.9 57%	
		In situ catalyst zeolite impregnation 5% Ni	4.6–3.5 23.9%	
Corylus avellana shells syngas	Fluidized bed reactor	Steam reforming 5% Ni-mayenite at 700 °C	27.7–8.17 69%	[82]
		Steam reforming 5% Ni-mayenite at 800 °C	27.7–4.1 86%	
		Scrubber with food waste oil as a fluid absorber	18.4 33%	
		Scrubber with food water as a fluid absorber	9.3 67%	

Table 2. Cont.

Sample	Process	Cleaning/Removal Method	Tar (g/Nm ³) BC/AF Efficiency	Ref
Cotton stalk	Steam gasification Fluidized bed reactor T: 600 °C S/B: 0.9	Olivine as bed material	452.07	[83]
	T: 800 °C S/B: 0.9	Olivine as bed material	19.7	
	T: 700 °C S/B: 0.9	Olivine as bed material	52.14–39.2 24% (compared to quartz sand)	
	T: 700 °C S/B: 0.9	Dolomite as bed material	52.14–31.34 39% (compared to quartz sand)	
Crushed almond shells	Steam gasification Bubbling fluidized bed reactor T: 830 °C S/B: 0.9	In situ catalytic ceramic filter candle with olivine as bed	0.72	[84]
	T: 830 °C S/B: 1.4		0.92	
	T: 830 °C S/B: 0.9	In situ commercial filter candle olivine as bed	1.9	
Pine wood sawdust	Air/Steam gasification Bubbling fluidized bed reactor T: 610 °C S/B: 3.41	In situ limestone calcinated at 900 °C as bed materials	83	[85]
	T: 650 °C S/B: 3.41		26.7	
	T: 700 °C S/B: 3.41		8	
	T: 610 °C S/B: 3.41	In situ silica sand and limestone calcinated at 900 °C as bed materials	62	
Naphthalene, toluene, and thiophene	Steam reforming laboratory-scale packed bed reactor T: 700 °C	Catalyst 31% NiO-40% MgO-30% Al ₂ O ₃	100%	[86]
	T: 750 °C			
	T: 800 °C			
	T: 700 °C	Commercial Ni-catalyst	13–4.11 70%	
	T: 750 °C		13–2.74 80%	
	T: 800 °C		13–1.09 92%	
Toluene	Gliding arc discharge reactor	Plasma reforming 3% CO ₂	16.1–6.1 62%	[87]
		Plasma reforming 9% CO ₂	16.1–6.6 59%	

Table 2. Cont.

Sample	Process	Cleaning/Removal Method	Tar (g/Nm ³) BC/AF Efficiency	Ref
Egusi melon shell	Air/steam gasification Fixed bed reactor T: 700 °C S/B: 1	0.2wt% Ni/dolomite/MgO / Al ₂ O ₃ dolomite calcinated at 850 °C	32–17 47%	[88]
	T: 800 °C S/B: 1		32–16 50%	
	T: 900 °C S/B: 1		32–15 54%	
	T: 1000 °C S/B: 1		32–14 57%	
Pine sawdust and bituminous coal	Steam gasification Decoupled triple-bed gasification T: 850 °C S/B: 1.3	In situ olivine calcinated at 900 °C	25.35–5.87 76% (compared to quartz)	[69]
		In situ olivine calcinated at 900 °C 5wt% Fe ₂ O ₃ /olivine	25.35–4.87 80% (compared to quartz)	
Sugarcane bagasse	Air/steam gasification Downdraft T: 843 °C ER: 0.3 S/B: 0.24	Cyclone separator	4.4–1.7 60%	[70]
		Wet scrubber	1.7–0.75 54.2%	
		Biomass filter	0.75–0.3 56%	
		Auxiliary filter	0.3–0.2 45.7%	
Pine sawdust	Catalyst Reforming Fixed-bed downstream T: 650 °C ER: 0.35	Pyrolysis char	43.9–9.3	[89]
		Natural dolomite	43.9–11	
		NiMo/Al	43.9–11.8	
		Fe/activated carbon	43.9–16	
	T: 750 °C ER: 0.45	Pyrolysis char	28.8–12.1	
		Natural dolomite	28.8–11.7	
		NiMo/Al	28.8–1	
		Fe/activated carbon	28.8–11.4	
	T: 750 °C ER: 0.45 S/B: 0.006	Pyrolysis char	15.9–3.4	
		Natural dolomite	15.9–4.8	
		NiMo/Al	15.9–5.2	
		Fe/activated carbon	15.9–0.6	

BC: before cleaning; AC: after cleaning; T: temperature; S/B: steam-to-biomass ratio; ER: equivalence ratio.

4.3.1. Biomass Gasification Parameters

The quality and yield of synthesis gas, as well as the concentrations of impurities, are influenced by several factors. The formation and consumption of each product inside the reactor are related to how the combustion and reduction reactions advance. They are a result of the lignocellulose composition in raw biomass, the catalytic effect of inorganics, and how all of these relations are influenced by reactor type and operational conditions:

temperature, reactor type, residence time, oxidizing agent (type and flow rate), biomass properties (feed rate, particle size, moisture content, and chemical composition), and pressure [52,90]. Therefore, the different combinations of biomass, gasifier design, oxidant agent, and other operational conditions lead to different gas compositions, impurities, and overall process costs.

Temperature

Temperature is one of the main parameters influencing gas distribution, tar formation, yield, and composition. At lower gasification temperatures (650–700 °C), primary tar is predominant, and the general concentration of tar is found to be very high. When temperature increases, tar undergoes cracking and reforming reactions, reducing its content while generating more permanent gases [64]. For example, when the temperature changed from 600 °C to 700 °C and 800 °C during experimental gasification in a bubbling fluidized bed (BFB) reactor, tar reduction was observed from 452.07 g/Nm³ to 39.2 and 19.72 g/Nm³ [83]. Tar decreased from more than 1 g/Nm³ at 900 °C to less than 0.2 g/Nm³ when it reached 1300 °C [91].

Zhou et al. [63] studied the air gasification of *Camellia sinensis* branches cellulose [52,91].

Regarding the influence of temperature in the tar profile, it has been reported that secondary and alkyl tertiary tars tend to decrease at higher temperatures and form more saturated compounds. In contrast, saturated tertiary tar has high thermal stability and tends to persist without decomposing. According to Tursun et al. [76], tar composition shifts from phenolic compounds and alkyl-substituted PAHs to non-substituted PAHs when temperature increases. The one-ring aromatics and phenols at 700 °C can be fully decomposed at 800 °C. The authors also found that naphthalene is the predominant tar compound in all temperature ranges, proving difficult to eliminate even under severe conditions.

Oxidizing Agent

The selection of the oxidizing medium for gasification significantly influences tar formation and gas composition. Typical agents are air, steam, O₂, CO₂, and their mixture. Air is by far the most used oxidant agent because of its low cost, stable operation, and abundance. However, air gasification produces a synthesis gas diluted in nitrogen (up to 51.9–55.4 vol%), resulting in low caloric value (3.5–6 MJ/Nm³ dry gas), poor hydrogen content, and a large proportion of char and tar [74]. CO₂ gasification has drawn attention to replacing N₂ as carrier gas because it can act as an inert agent at low temperatures. When heated, CO₂ can react with char in Boudouard reaction and tar in dry reforming. Therefore, CO₂ can increase the process's cold gas efficiency and carbon conversion [75].

A significant parameter that affects air gasification is the equivalence ratio (ER), which is the ratio of the real oxygen supply to fuel divided by the oxygen needed for the complete combustion of theoretical biomass to fuel. ER indirectly affects the reactor's temperatures and pressures [92]. ER has an optimal range value reported to be between 0.2 and 0.4. Lower ER results in incomplete gasification and excessive char and tar formation, and higher ER produces more non-combustible gases (CO₂ and H₂O) at the expense of H₂, CO, and CH₄. Gai and Dong [93] observed during the air gasification of corn stalks in a downdraft reactor a continuous decrease of tar content from 7.2 g/Nm³ to 0.41 g/Nm³ by raising ER from 0.18 to 0.41. Varying ER from 0.18 to 0.32, H₂ and CO increased from 6.91 to 13.51 vol% and 11.35 to 19.81 vol%, respectively. Meanwhile, for higher ER, both had a slight decrease.

Oxygen results in syngas with a medium to high caloric value (10–18 MJ/Nm³) and improved quality in terms of syngas composition and the presence of tar. It is reported to produce the lowest tar and char content because it promotes the secondary cracking of

tar and endothermic reactions. Pure oxygen is not a preferable gasification agent because it is not cost-competitive, limiting wide-range applications. For commercial applications, some research has explored gasification in an oxygen-rich atmosphere. Under an oxygen-enriched air condition and ER constant, the increase of oxygen concentration in carrier gas enhances the oxidation reaction. It was observed that O_2 concentration from 21 to 31.4 vol% decreased tar content from 135 to 100 g/Nm³, and H_2 and CO increased by 65 and 23%, respectively [94]. According to Zhao et al. [95], for every 5% increase in oxygen concentration, the volume of gas production reduces by 12–17%, and heat value and hydrogen concentration increase by about 14–18% and 11–14%, in this order. Excessive oxygen in the carrier gas can be problematic because the air volume is too low, making fluidization difficult.

Using steam as an oxidant agent is an effective medium for producing high-quality gas and higher hydrogen yields. Steam favors the water/gas reaction, water/gas shift reaction, and steam/methane reforming reaction, offering a higher yield of H_2 , a greater H_2 /CO ratio, and decreasing tar concentration [83]. Although more competitive on an industrial scale than oxygen, steam requires an external energy supply to be heated. To reduce operational costs, some studies have investigated the role of air/steam gasification as a more economically viable technology [96]. In experiments by Cerone et al. [97], the authors obtained an increase in the H_2 content from 9.7–11.4% in air gasification to 15.8–21.6% with the addition of steam. Similarly, Nakyai et al. [98] improved hydrogen concentration from 23.26 to 46.82 mol/kg of biomass and energy efficiency from 66 to 85% upon steam injection. Nonetheless, excess steam may reduce the overall gas yield and carbon conversion efficiency, which can increase tar content.

The steam-to-biomass ratio (S/B) determines steam or air/steam gasification performance. The literature has reported that it is necessary to maintain an S/B above at least 0.7. For optimum gasification efficiency, the S/B is suggested to be between 0.9 and 1.2. Increasing S/B is reflected in increasing the steam concentration and partial pressure, which corresponds to Le Chatelier's principle. However, once the carbon conversion is complete, the excess steam has a negative impact [83]. Yisheng et al. [99], varying S/B from 1.1 to 1.8, observed a dry gas yield increase from 0.88 to 1.18 Nm³/kg and an enhanced tar cracking reaction, which caused a reduction in tar yield. Matthias et al. [100] and Tian et al. [101] reported the minimum total tar content of 5.02 and 4.2 g/Nm³ when using the S/B ratio of 1–1.2.

When concerned with gasification medium, the general order for obtaining the highest amount of H_2 is reported as steam > oxygen > air > CO₂ [102]. In summary, pure steam or oxygen as a gasifying agent is great for increasing the yield of combustible gases and producing less tar, but a mixture of different agents can be preferable to combine performance and economic viability.

Gasifier Design

Several gasifiers have been used to conduct biomass thermochemical conversion. The most common reactor types are fixed bed (updraft, downdraft, and crossflow), fluidized bed (bubbling and circulating), and entrained flow [7]. Fixed bed gasifiers have been widely used because they are simple to operate and require less maintenance. Those are classified into updraft, when biomass enters from the top and oxidizes from the bottom, and their interaction is counter-current; downdraft, when both gasification agent and biomass flow co-current; and crossflow, when material flows downward, and the air is supplied from both sides. Crossflow gasifiers are less investigated in biomass gasification because they provide high tar content and low gas production efficiency [103].

Updraft gasifiers are the most straightforward and oldest form of gasifiers. The produced gases, tar, and other volatile compounds are dispersed at the top of the reactor, while ash is removed at the bottom [104]. It offers the advantages of using high moisture biomass with high efficiency. However, a major drawback is the high level of ashes and tar in the outflow gas because of the lower outlet temperature [105]. Downdraft reactors provide higher yields of hydrogen and reduced tar content due to the gas flow directions and solid matter enhancing the gas/solid interactions and continuously cracking through the high-temperature oxidation zone at the gas outlet [106]. It is attractive for biomass gasification because of the quality of gas and easy operation, but it can be restricted for small applications (10 kW–1 MW) because the thermal nonuniformity makes it difficult to scale up. Additionally, it may have limitations for low bulk density feed and high moisture content [107].

In a fluidized bed gasifier, solid particles circulate within the bed due to the action of circulating gas, which prevents issues such as solid accumulation and coking. The gas-solid two-phase flow promotes the mixing of solid particles, leading to improved uniformity of reactions and compositional uniformity of the produced syngas [108]. The fluidized bed has a fast gasification rate and a higher production rate of H_2 and CO. The major problem is related to the high content of ashes and the complex construction and operation costs compared to fixed beds. Nonetheless, it is a widely used and researched technology because it allows operating in medium and large gasification plants. Based on the flow pattern and combination features, they can be classified as bubbling fluidized beds (BFB), circulating fluidized beds (CFB), and dual fluidized beds (DFB) [109].

In BFB, biomass is fed from the side into the hot moving bed fine-grained material and is maintained in a fluidized state flowing upward with the gasifying agent, producing volatiles and char. BFB has a uniform temperature, is flexible to feed, and tends to produce more CO_2 and CH_4 than syngas because of the low temperature in the top section [110]. CFB consists of a riser where fuel undergoes drying and pyrolysis, releasing volatiles, and a high-temperature cyclone separator that separates product gas from remaining solids, which return to oxidizing with the bed [111]. The CFB design led to high velocities of the recirculation system and violent mixing, providing excellent heat transfer that improves the feedstock's reaction rate and conversion efficiency and offers better gas/solid mixing. Despite that, operational plants are complex and require high capital investment [103].

A DFB is an allothermal type of gasification system consisting of a combustion furnace and gasifier interconnected through a circulating bed material but controlled separately [112]. Standard configurations involve (1) a bubbling fluidized bed (BFB) gasifier fluidized with steam, which converts biomass into raw syngas gas/product, and (2) a CFB or fast fluidized bed combustor (riser), which oxidizes the residual char in the presence of an oxidizing (usually air) or a fluidizing agent providing heat for the highly endothermic gasification reactions [109]. The main advantage of DBF is the separation of gasification and combustion reactions, which enables the use of air in the combustor without the effect of nitrogen dilution. Syngas heating values and hydrogen content of allothermal gasifiers are comparable to gasification conducted with steam or oxygen. The gas production by the gasifier is primarily composed of H_2 , CO, CO_2 , and CH_4 , with only small amounts of tar and other undesired components [109].

An entrained flow reactor works on a co-current basis, with a high gasification temperature (up to 1500 °C) and a short residence time. It produces very low CH_4 and CO_2 , with a high content of H_2 and CO and almost complete carbon conversion, and no tar in the outgoing gas, often considered a tar-free gasifier [113]. Entrained flow gasification has shown the best syngas quality among existing gasification technologies for high-grade biofuel synthesis. Despite that, it requires homogeneous and high-density feedstock fuel

(usually in powdered form); therefore, most of the uses of entrained flow gasifiers remain focused on coal gasification, whereas for biomass gasification, there are some technical challenges such as ash melting [114,115].

To overcome some drawbacks of biomass gasification, researchers worldwide have performed and reported various modifications of the basic design of gasifiers. Design improvements can be made by modifying the feeding, air supply, producer gas recirculation, and discharge systems [107]. Omar and Munir [116] proposed optimizing the downdraft gasifier by inserting nozzles in the firebox to maintain uniform air distribution for smooth and trouble-free operation of corn cob gasification during power generation. Structure changes directly increased the reactor's internal temperature, which minimized tar production (3.2 g/Nm^3) and enhanced producer gas composition (8.43 MJ/Nm^3). The authors also found that pre-heating of the oxidizing agent provides minimal tar production because it increases the carbon conversion efficiency.

Kumar et al. [117] proposed a new dual-stage biomass gasification equipped with a downdraft reactor using air as an oxidant agent and acacia wood as feed. A 213 mg/Nm^3 tar concentration was measured at the double-stage gasifier outlet. Then, after cleaning and cooling units in the regenerator, fabric filter, air cooler, water cooler, and paper filter, tar content was 100 mg/Nm^3 , 52 mg/Nm^3 , 34 mg/Nm^3 , 20 mg/Nm^3 , and 8 mg/Nm^3 , respectively, which was considered suitable for an internal combustion engine. The tar content in single-stage gasification was found to be 72 mg/Nm^3 after the paper filter.

Wang et al. [118] proposed for the steam/oxygen gasification of pine chips a novel two-stage gasifier by coupling a downer CFB reactor for biomass pyrolysis (high mass and heat transfer) and a riser downdraft reactor (high tar removal rate) for char gasification and tar cracking. As a result, the long-term running at ER 0.35, S/B 0.15, and temperature of 950°C led to the tar content of 95.5 mg/Nm^3 .

Khan et al. [119] investigated a flexible upscaling of fixed bed systems by supplying heat through an updraft flue gas heating jacket. The authors found the optimum injector configuration at the reactor top, and then tar contamination in the produced gas ranged from 13.63 g/m^3 at the lowest S/B ratio down to 7.38 g/m^3 at the highest S/B. Rahman et al. [120] designed a low-tar biomass gasifier based on a downdraft reactor. A separate combustor is installed inside the reactor in the partial oxidation zone to increase the mixing of the gasifying agent with the pyrolysis gas. This configuration achieved a tar content of $10.6\text{--}29.8 \text{ mg/Nm}^3$ in the producer gas using air as an oxidant.

4.3.2. Thermal Cracking

As explained in Section 4.3.1, temperature is one of the most important parameters that affects the gasification process. Thermal cracking is an intensive method that uses the effect of high temperature (above 1000°C) to crack tar molecules into lighter gases. It results in a decrease in tar content and an improvement in gas quality and yield [121]. In a two-stage fixed bed reactor, Zhai et al. [122] performed the secondary thermal cracking of rice husk tar. At 1200°C and a residence time of 0.5 s, tar yielded 11.7 mg/Nm^3 , and no tar was collected at 1300°C . Because of the severe condition (high temperature and short residence time), in this process, the tar cracking rate could reach 99.98%; however, it also implies the generation of a large amount of macromolecular compounds and soot precursors and a high operating cost. Instead of thermal cracking, catalytic reforming has been considered more economical and technically viable for gas purification and tar reduction [121].

4.3.3. Gasification Catalyst

Catalysts are among the most efficient and convenient ways to remove or reduce tar content. They allow for conducting gasification at low temperatures (500–700 °C) and boost heat and mass transfer. Catalysts improve hydrocarbon reactions such as thermal cracking, steam and methane reforming, dry reforming, and water/gas shift reactions, converting tar into valuable fuel gases (H_2 , CO, CH_4 , and CO_2) [123]. Compared to thermal cracking, tar from catalytic reforming has more light compounds [121].

Catalytic tar conversion can be performed in in situ or ex situ mode. In situ occurs in the gasifier during gasification by wet impregnation, dry mixing, or as bed material in a fluidized bed reactor. Ex situ mode employs a secondary downstream reactor for tar removal in post-gasification. The catalytic reactor operates under different conditions of the gasifier unit. Secondary methods may be more efficient than primary but can be more complex for scaling up and require higher operating costs [124].

Different types of materials can be used as catalysts, which can be broadly categorized into (i) natural catalysts (dolomite, olivine, calcium oxide), (ii) alkali (K, Na, and Li), and alkaline earth (Ca and Mg) metals, (iii) transition metals (Ni/Fe/Co), and (iv) carbon-based catalysts. Each material has different catalytic activity depending on its origin and characteristics as well as the gasification conditions. The main success of cracking catalysis is based on the proper selection of catalytic materials that should present high efficiency on tar removal at temperatures of 600–900 °C; the capability of providing a high yield of desired products (mainly syngas); high mechanical stability and resistance to poisoning; easy regeneration; low cost; and commercial availability. The catalyst selection also depends on whether it is used for in situ tar cracking or post-gasification treatment. Natural catalysts and alkalis are generally preferred as primary catalysts. Ni-based is best employed as a secondary catalyst because it reduces deactivation problems by coke formation on the catalyst surface [124].

Natural Catalyst

Natural minerals such as dolomite, olivine, and calcinated rocks (limestone and magnesite) are widely used as catalysts in biomass thermochemical conversion. These minerals are active for biomass tar cracking, inexpensive, found in abundance, and can be used directly or after some pretreatment [125]. Major problems of natural catalysts are susceptibility to deactivation due to coke formation and carbon deposition and low thermal stability and mechanical strength, especially when compared to synthetic ones. Natural mineral characteristics can be improved through calcination, which helps in chemical activity and mechanical strength, and by loading active metals (like Fe, Ni, Co, and Ce).

Dolomite and olivine are often used as bed material in fluidized bed reactors, while CaO's primary application is as a sorbent for CO_2 in situ capture [126]. Dolomite is a carbonate mineral composed of CaO (66.28%), MgO (32%), and other minerals. It has shown the best catalytic activity among natural minerals, being effective in tar reduction and conversion and increasing syngas yield, H_2 /CO ratio, and HHV. CaO is active in tar cracking, while MgO helps inhibit stable carbon formation at the CaO surface. In addition, iron's presence in dolomite plays a role in the steam reforming of tar [127]. In experiments by Al-Obaidi et al. [128], the use of dolomite as a primary and secondary catalyst at 850 °C reduced tar content by 78% and 99%. Followed by downstream gasification, a 20% increment in gas yield and 66% in H_2 were also reported.

Dolomite activity is greatly enhanced when in calcinated form because oxides can eliminate tar by steam and dry (CO_2) reforming reaction. Even calcinated dolomites are prone to deactivation by carbon deposition, abrasion, elutriation, and sintering, resulting in continuous strength reduction over time [129].

However, calcination increases pore volume and diameter, providing better mass transfer and preventing cake formation (sintering). During air gasification of rice husk in a fluidized bed reactor at ER 0.14 and 950 °C, the calcinated dolomite reduced tar content by 88.1% compared to 73.02% using natural dolomite [130]. Similarly, Cao et al. [129] evaluated the increasing calcined dolomite concentration in the bed for air gasification of pine sawdust. This resulted in high H₂ and CO content, whereas the CO₂ and CH₄ content declined because of the enhancement of the methane-reforming process. At the temperature of 850 °C, an ER of 0.2, and a dolomite content of 50%, syngas were obtained with the maximum HHV (6.55 MJ/Nm³) and minimum tar yield of 4.6 g/Nm³. Under the non-catalyzed process, tar concentration was 11 g/Nm³.

Olivine comprises silicate minerals. Its catalytic activity for tar elimination can be related to the magnesite (MgO) and iron oxide (Fe₂O₃) content. It is more effective for light tar than heavy ones, and compared to dolomite, olivine is reported to have poor activity [131]. According to Yang et al. [83], during catalytic gasification of cotton stalk, the olivine presence resulted in 31.45% of hydrogen and 31 g/Nm³ of tar. Under the same conditions, olivine resulted in 23.72% of H₂ and 39.2 g/Nm³ of tar, which were comparable to the performance of quartz sand (22.79% of H₂ and 52.14 g/Nm³ of tar) that has no catalytic activity. Similar results were observed in fluidized bed gasification with different bed materials conducted in a bench-BFB reactor using air/steam at 850 °C. Olivine behaved similarly to sand, and the activity of the bed materials decreased as dolomite > MgO > olivine B/kaolin > sand, corresponding to tar concentrations of 0.48, 0.63, 1.63, and 3.63 g/Nm³ [132].

Olivine is interesting because it produces lower particulates, suffers less with coke deposition and attrition resistance, and has higher mechanical strength, equivalent to sand [133]. Due to abrasion resistance and mechanical properties, olivine is suitable as a circulating fluidized bed material. Its performance can be improved by loading active metals [131]. When loaded with Ni-Fe bimetallic by wet impregnation and thermal infusion, olivine catalyst reduced tar content by 87–89% and 78–82% compared to silica sand and raw olivine as bed material in CFC [114].

Calcium oxide contains minerals like limestone and dolomite. It can be obtained from pure CaCO₃, Ca(OH)₂, and calcium acetate. It is interesting because it exhibits good tar cracking activity, depending on the precursor, and acts as a CO₂ sorbent [124]. It has been introduced in novel gasification processes such as sorption-enhanced gasification (SEG) and CaO-based chemical-looping gasification (CLG). In CLG, an oxygen carrier (typically metal-oxides) is used to transport the lattice oxygen from the air to the fuel, supplying the oxygen needed for gasification without contact with biomass and air. In SEG, a CO₂ sorbent (CaO) is used in situ in a steam dual fluidized bed reactor. CaO captures CO₂ and forms CaCO₃, shifting the equilibrium of the water gas to hydrogen. The integration of calcium chemical looping and SEG, known as a sorption-enhanced chemical looping gasification (SECLG), has been proposed for the combined performance of CaO as the CO₂ sorbent and metal oxides. The combination lowers the energy penalty and operating cost regarding the [115,134] reaction temperature. At 670 °C, CaO/B decreased tar yield from 15.07 g/Nm³ to 6.6 g/Nm³ when increasing the CaO/B ratio from 0.0 to 2.0 [135].

Overall, natural mineral catalysts are challenging because of their fragile nature and low activity and stability. Compared to synthetic metal-based catalysts, which can almost completely remove tar, for most gas applications, mineral catalysts do not reach the low levels of tar required [136]. Although such minerals are active for the conversion of biomass tar into syngas, pretreatment and modifications are considered indispensable for highly efficient reduction [137].

Akali and Alkaline Earth

Akali, alkaline earth (AAEM), and transition metals are the most commonly used catalysts. AAEMs have been used as cost-effective catalysts for the thermochemical conversion of carbonaceous materials. They are natural, widely available, price-competitive, provide high reforming efficiency, and have good resistance to carbon deposition [138]. Considerable amounts of AAEM species occur naturally in lignocellulosic biomass structures and are known to influence the kinetics of gasification reactions [139]. The presence of intrinsic AAEMs in biomass enhances the heterogeneous char gasifications and the homogeneous WGS, steam/tar reforming, and hydrocarbon reforming reactions. Then, less tar is produced during the thermoconversion process [139].

Mineral oxides (CaO , K_2O , MgO , and Na_2O) or carbonates (Na_2CO_3 and K_2CO_3) are catalytically active forms of AAEM precursors. Sodium and potassium titanates have been introduced as novel alkali carriers to replace carbonate because of their high melting point and stability for ex situ reformers. About 99% of tar (from 208.8 g/Nm^3 to 0.93 g/Nm^3) can be converted under $4\text{Na}_2\text{O}\cdot 5\text{TiO}_2$ reforming [140].

Alkali metals tend to be more active than alkaline, and catalytic activity has been reported to decrease in the order of K, Na, Ca, and Mg [141]. The limited catalytic effect of Ca and Mg is related to their low diffusivity, sintering activity, and poor selectivity [138]. The major interest in CaO as a catalyst is its capability to capture CO_2 and absorb other unwanted species, such as sulfur. Otherwise, potassium is catalytically active even at small concentrations, especially for heavier compounds. Bach-Oller et al. [142] showed that the impregnation of biomass with K creates a gas-phase-induced phenomenon. Potassium is volatilized from the ash-covered bed material and is transported to the char through the gas phase. With and without direct contact with the biomass, K presence significantly reduced the yields and molecular size of gas impurities (tar and soot) and the amount of unconverted carbon. According to Berguerand and Vilches [143], the gasification with alkaline feldspar $[(\text{K}, \text{Na})\text{AlSi}_3\text{O}_8]$ led to a total tar level lower than the operation with quartz sand and with similar species distribution when using olivine as bed material.

Sodium in the gas phase promotes volatile reformation and tar cracking, demonstrating a significant catalytic effect on lignin gasification. Impregnation with Na_2CO_3 , NaHCO_3 , and Na_2SO_4 can increase the content of H_2 from 10 mmol/g to $40\text{--}75 \text{ mmol/g}$ while decreasing tar and char formation. Nonetheless, AAEMs are reported to have ash-enhanced catalytic effects and are often related to slagging and agglomeration issues [139]. Additionally, K and Na react with aluminum and silicon present in the feed to form stable and unreactive silicates and aluminosilicates.

Transition Metals

Noble metal catalysts like Pt, Rh, and Ru show excellent tar reforming. They have high resistance to impurities such as sulfides and robustness to coke deposition [144]. The performance in tar reduction is observed as $\text{Rh} > \text{Pt} > \text{Pd} > \text{Ni} = \text{Ru}$ [60]. Nonetheless, the higher cost hinders noble metals from being used in large-scale systems, and they are mostly used as promoters in nickel-based catalysts [136]. Other transition metals such as Co, Fe, Zn, Mo, and Cu have demonstrated a promising ability to enhance the quality of produced syngas. Mo-based has shown strong oxygen adsorption for deoxygenation and rich-opening capacity for oxygen-containing macromolecules [145].

Cobalt is the preferred catalyst for Fischer–Tropsch synthesis. Co may not contribute directly to the hydrogen formation but strongly influences CO_2 adsorption. It promotes the Boudouard reaction and consumes the coke deposited on the surface to produce more CO, conferring high resistance to carbon deposition [146]. Toluene conversion rates were

reported to increase from 5 to 20% under catalyst-free and biochar to around 70% when using biochar-supported cobalt-based catalysts [147].

Fe-based catalysts have poor reactivity and are more prone to oxidation, which decreases activity. Nonetheless, they are economically more attractive and non-toxic, promoting the decomposition of large aromatic rings, steam reforming, and a shift in water/gas. Various iron-based materials have been explored for tar elimination and have gained growing interest as alternatives to nickel and other metal catalysts. Iron-based catalysts express different levels of activity based on their oxidation state (Fe_2O_3 , Fe_3O_4 , FeO , or Fe^0). The form of metallic iron (Fe^0) can break C-C and C-H bonds and is suggested as more catalytically active for tar and methane conversion than iron oxides [148].

Khajeh et al. [116] reported a total tar removal efficiency of 84.20% using the iron catalyst supported on the activated carbon. Shen et al. [149] reported a heavy tar conversion of 42, 82.8, 92.3, and 93% when mixing biomass with char, Fe-char, Ni-Fe/char, and Ni-char. As expected, monometallic Ni catalysts exhibited much higher reforming activity. However, the bimetallic Ni-Fe char-supported catalysts reached comparable efficiency. Bimetallic Ni-Fe has cheaper synthesis because of the low concentration of Ni. Zhang et al. [145] proposed a biochar-supported Fe-Mo carbides catalyst. They observed a high tar conversion efficiency of 91.05% in ex situ configuration at 700 °C. The bimetallic catalyst inhibited the formation of multiring tar compounds, removed unstable oxygen-containing functional groups, and exhibited high cycling stability.

Ni-based catalysts are the commercial catalysts most investigated for both primary tar removal and secondary gas reforming post-gasification. They are mainly prepared from the precursor $\text{Ni}(\text{NO}_3)_2$ [123,126,150]. Ni catalyst is more expensive than alkaline and other transition metals but is highly active in converting heavier hydrocarbons, methane reforming, and WGS reaction [151]. When used as the secondary catalysts, the supported nickel catalysts can reach complete tar decomposition [137]. Most problems related to Ni-based catalysts are attrition, rapid deactivation by coke deposition, metal sintering, and sulfur and chloride poisoning [152]. Some limitations of nickel and other metal catalysts can be avoided by selecting suitable supports, forming alloys, and modifying by adding a promoter (K, Na, Li, Mg, Fe, Cu, Cr, La, Co, and Mo).

The catalytic support provides active sites for loading metals and prevents the leaching of active metals. Various materials can be used as support, such as zeolites, perovskite, dolomite, olivine metal oxides (CeO_2 , SiO_2 , ZrO_2 , TiO_2 , MgO , ZnO , La_2O_3 and Al_2O_3), and carbon (activated charcoal) [82]. Supports must be selected based on their acidity/basicity, specific surface area, pore structure, and electronic properties. Pore structure directly affects the heat and mass transfer, which is decisive in determining the rate of gas/solid reactions. High surface area is beneficial for providing more availability of catalytic sites; otherwise, higher surface acidity increases the tendency of carbon deposition and lowers [153,154].

For nickel, γ -alumina (Al_2O_3) is the commercially used support. It is cheap and has a high surface area and mechanical strength, providing better Ni dispersion. Vivanpatarakij et al. [137] investigated the effect of Ni supported on Al_2O_3 , CaO , and MgO in the steam reforming of model compounds of tar, C_{10}H_8 , C_7H_8 , $\text{C}_6\text{H}_6\text{O}$, and $\text{C}_{10}\text{H}_{10}$. In their experiments, Ni/ Al_2O_3 showed superior reactivity compared to Ni- CaO or Ni- MgO , and 20% Ni/ Al_2O_3 showed the highest H_2 concentration and tar conversion compared to 10% and 15% Ni.

Because γ -alumina has a limited lifetime and high coke formation rate, studies have examined the addition of metals and metal oxide promoters to improve stability [155,156]. Mazumder and Lasa [155] modified Ni/ Al_2O_3 catalysts using La_2O_3 as a promoter for the steam gasification of biomass. La_2O_3 reduced the alumina support's thermal sintering and acidity, and the developed catalyst performed close to the thermodynamic equilibrium.

Claude et al. [157] found the optimal catalytic activity of Ni/ γ -Al₂O₃ catalysts when combined with two different metals, Mn + Mo or Co + Mo. They provided a balance between a high toluene reforming rate and low amounts of carbon. In a staged rice husk gasification, the catalyst Ce–Ni/ γ -Al₂O₃ showed stability to maintain the H₂ concentration above 67.21 vol% during 5-lifetime tests. Santamaria et al. [158] modified Ni/Al₂O₃ catalyst by incorporating CeO₂ and MgO. MgO led to faster catalyst deactivation. CeO₂ favored coke gasification and oxidation, showing a lower average coke deposition rate. Vecchione et al. [82] proposed a study concerning the removal of tar at low temperatures. They placed a catalyst consisting of 5% nickel and 95% mayenite downstream of the particulate removal system. Tar was found to be mainly composed of class 5 light aromatic hydrocarbons characterized by a single ring (including toluene, benzene, xylene, and styrene). Regarding the catalyst effect, the efficiency of tar compound removal at 700 °C was more than 80%. At 800 °C, most of the compounds were above 90%.

Promoters are considered the most critical modification strategy to achieve high catalytic activity and stability at low temperatures. They reduce the problems with coke formation and Brønsted acid centers. The synergetic effect between bimetallic active sites optimizes the adsorption of target gases and redox properties [145,150]; noble metals and AAEMs (K, Ca, and Mg) are alloys as promoters. AAEM and rare earth oxides are great for providing more oxygen vacancies and active oxygen mobility. They improve oxygen adsorption on or around Ni's surface, which is beneficial for the WGS reaction [159].

The interaction of Ni-Fe is found to have superior activity and stability compared to monometallic catalysts. Zhang et al. [160] observed that the activity and stability of Ni-char-based catalysts are significantly improved by the introduction of Fe as a co-catalyst. Toluene conversion was 95.6% with Ni-Fe/char, compared to 91.8% with Ni/char. In catalytic cracking, Yang et al. [161] employed biochar–(Fe 2.5%)-(Ni 2.5%). Toluene removal was 92.76%, higher than biochar-Ni 5% (89.48%). Huang et al. [152] investigated Ni-based catalysts alloyed with Cu. Compared to Ni or Cu catalysts, Ni–Cu/biochar was more effective in tar removal (93.2% at 800 °C) and syngas production. The bimetallic showed resistance to sintering and coke deposition (coke amount about 48 mg/g compared to 313 mg/g using Ni/biochar). Complete elimination of tar was obtained by Lysne et al. [144] with noble metal-promoted (Pt, Pd, and Rh) Ni-Co/Mg(Al)O catalysts.

In addition to metal type, the precursors and the particle size of metal also significantly affect performance. In general, small particle size results in better catalytic activity. Therefore, selecting the preparation method to achieve ideal properties is also necessary [159]. The easiest preparation method is impregnation, but active metals tend to agglomerate on the surface. Alternative methods include calcination methods [149], one-step pyrolysis [152], and microwave firing [162]. One-pot hydrothermal, one-step pyrolysis, and ion exchange can be used for uniform load when using carbon materials as support [152].

Zeolites

Zeolites are aluminosilicate materials used as commercial catalysts in several frameworks, such as Y, XY, ZSM5, and F9, most of which do not occur naturally [163]. Zeolites have a large specific area, intense surface acidity, and high thermal and hydrothermal stability. They are resistant to sulfides, easily regenerate, and provide high tar reforming activity [136].

Waluyo et al. [81] modified mordenite zeolite by acid leaching and used it as a catalyst in the steam gasification of palm kernel shells at 750 °C. Syngas was produced with a tar content of 0.12 g/Nm³ and H₂/CO ratio from 2.7 to 5.7. El-Rub et al. [163] investigated the effectiveness of natural Jordanian zeolite as a catalyst for toluene (as a tar model compound) in thermal cracking and catalytic dry reforming reactions. For the thermal cracking of

toluene at temperatures from 700 to 900 °C, the formation of methane, ethane, m-xylene, and benzene was observed. The highest tar conversion of 90% was observed at 900 °C, compared to 13% at 700 °C. Authors also reported that zeolite's color changed from brown to black in temperatures >800 °C, indicating deactivation by coke production as an effect of acidity.

Under temperatures above 800 °C, toluene reacts with carbon dioxide to form carbon monoxide in the reforming reactions on zeolite's surface. Hydrogen gas structure rearranges and recrystallizes, hindering its catalytic activity. Zeolites are more prone to cracking of tar than to engage in reforming reactions because of the Brønsted acid sites in the pores of zeolite's structure. Essential parameters of zeolite performance are its contained ions that create Brønsted acid sites and its pore structure that creates a sieve-like structure with small pore sizes that limit the diffusion of molecules [137].

Carbon-Based Materials

Carbon-based (C-based) materials have been extensively investigated and applied as catalyst supports in various industrial and environmental applications due to their chemical stability, high specific surface area, and low cost. Their micropore structure and surface area make them a preferred adsorbent for tar elimination, converting tar into lighter gaseous hydrocarbons.

Kim et al. [1] prepared Ni-based catalysts using three different carbon supports: activated carbon (AC), carbon black (CB), and low-rank coal (LRC). They evaluated the catalytic activities of NiMg/CB, NiMg/AC, and NiMg/LRC for steam reforming of 30 g/Nm³ toluene at 500 °C. The AC catalyst exhibited the highest carbon conversion at 95%, while the performances of NiMg/CB and NiMg/LRC were approximately 87% carbon conversion. The catalysts supported on AC and LRC were found to be unstable when exposed to steam at elevated temperatures, while NiMg/CB demonstrated high stability, maintaining consistent catalytic performance without any decline during 50 h of continuous steam reforming operation.

Cobalt-based catalysts utilizing lignite as a carbon precursor were synthesized through an economical and straightforward ion exchange method for the steam reforming of toluene. The catalyst achieves a maximum toluene conversion of 98.7% at 400 °C and shows high stability during the 100-h test [3].

Chars and biochars derived from coal or residual biomass are both inexpensive carbonaceous materials with similar physicochemical properties, primarily resulting from biomass thermochemical conversion processes. While char is not as effective as Ni-based catalysts in terms of catalytic activity, its higher porosity is suitable for tar adsorption and enhanced cracking. Additionally, char-supported catalysts are well known for their cost-efficient and high catalytic performance. In the work by Hu et al. [2], the authors managed to reduce tar content from 101.37 g/kg biomass to 6.46 g/kg biomass when switching from a char catalyst to a Ni-6%/char catalyst. Additionally, the effectiveness of catalysts in removing tar and enhancing H₂ content was ranked as biochar < coal char < Ni/Biochar < Ni/coal char [3].

Even though coal char has outperformed biochar as a catalyst, biochar is widely recognized among carbon-based materials as a promising option for addressing the challenges of tar reforming while also aligning with sustainability in production and environmental impact. The following Section 5 will discuss biochar-based catalysts and their advantages in contributing to a circular and sustainable solution for waste.. The tar conversion under different catalysts is summarized in Table 3.

Table 3. Summary of tar conversion under different catalysts.

Sample	Catalyst	Operational Conditions	Tar Conv. (%)	Tar Yield (g/Nm ³)	Ref.
Pine sawdust tar	Pine sawdust char	Fluidized bed reactor Downstream 750 °C	65.7		[125]
		950 °C	79.1		
Empty fruit bunch	Malaysian dolomite	Fixed bed reactor Downstream 900 °C	94–99%		[128]
Pine sawdust	10 wt% dolomite Calcined at 900 °C	Air gasification Fluidized bed reactor 700 °C		~9.5	[129]
	10 wt% Calcined at 900 °C	850 °C		~6.5	
	50 wt% dolomite Calcined at 900 °C	850 °C		4.6	
Rice husk	Dolomite Calcined at 830 °C	Air gasification Fluidized bed reactor 950 °C		1.8	[130]
	Raw dolomite	950 °C		4.1	
Cotton stalk	Olivine	Steam Fluidized bed reactor 600 °C		452.07	[83]
		800 °C		19.72	
		700 °C		39.2	
	Dolomite	700 °C		31.3	
Bark pellets	Dolomite	Air/steam Fluidized bed reactor 850 °C	87	0.48	[132]
	Olivine B/kaolin		54	1.67	
	MgO		83	0.63	
Pine sawdust	Olivine	Fluidized bed reactor Air 850 °C		4.49	[114]
	Olivine calcinated at 900 °C			2.14	
	Olivine calcinated at 1000 °C			1.98	
	5% Ni-Fe/olivine calcinated at 1100 °C			0.83	
	5% Ni-Fe/olivine calcinated at 1400 °C			0.77	
Wood sawdust	4Na ₂ O·5TiO ₂ /char	Reforming Fixed bed reactor 850 °C	99.6	0.93	[140]
	Na ₂ O·3TiO ₂ char		96	3.2	
Toluene	Co/biochar	Steam reformer Fixed bed reactor	62–70		[147]
	Co-Ni/biochar		95–97		
	Co-Fe/biochar		80–90		

Table 3. Cont.

Sample	Catalyst	Operational Conditions	Tar Conv. (%)	Tar Yield (g/Nm ³)	Ref.
Pinewood tar	Co-Ni/biochar	Steam reforming Fixed bed reactor 700 °C	91		[147]
Naphthalene	Reduced bauxite residue	Fixed bed reactor Steam reforming	55–90		[148]
	HCl-activated bauxite residue		55–97		
	Gasification biochar		50–85		
	Biochar-activated bauxite residue		95		
Rice husk tar	Ni-Fe-char	Reforming 800 °C Air	92.3		[149]
Aspen wood sawdust	Ni/char	Two-staged fixed bed reactor 700 °C	93		[152]
	Char		42		
	Fe/char		82.6		
	Biochar		55		
	Cu/ biochar		65		
	Ni/ biochar		80		
	0.5Ni-Cu/ biochar		82		
	Ni-Cu/ biochar		90		
	Ni-0.5Cu/biochar		87		
Toluene	10 wt% Ni/ γ -Al ₂ O ₃ dopped 2wt% Mo	650 °C	55		[164]
	10 wt% Ni/ γ -Al ₂ O ₃ dopped 2 wt% Mn		38		
	10 wt% Ni/ γ -Al ₂ O ₃		50		
Toluene	2.5% Fe-2.5% Ni/calcinated biochar	Catalytic cracking 800 °C	92.7		[165]
	5% Fe/calcinated biochar		88.5		
	5% Ni/calcinated biochar		89.4		
Biomass-derived tar	5% Ni-Fe-Ti	900 °C	84.6		[162]
	10% Ni-Fe-Ti		99		
Toluene	Natural zeolite	800 °C	63		[163]
		900 °C	79		
Spruce wood pellets	Biochar	875 °C Fluidized bed Steam downstream operation	64–70		[166]

Nanocatalyst

An approach that has been addressed to advance specific aspects of tar issues is the use of nanomaterials. Nanomaterials are composed of one or more compounds that are at least 1 nm to 100 nm in size. At this scale, the materials may have new physical and

chemical properties [1]. Nanotechnology has been associated with several fields, triggering significant advances in industrial processes.

Nanomaterials have garnered attention in biomass gasification due to their potential resistance to carbon deposition deactivation, high selectivity, and stable properties in tar removal. A low-cost nanocatalyst with anti-deactivation ability was prepared by Chen et al. [2] for the in situ conversion of tar. The catalyst was synthesized using pine sawdust biochar as a support, nickel as the active component, and Fe/Co/Mg as promoters. When compared to other studies in the literature, the proposed catalyst achieved the highest tar conversion of 99.59% and increased H₂ yield by more than 30% compared to traditional catalysts.

Yang et al. [3] showed the great benefits of biochar-based nanocatalysts loaded with Ni/Ca/Fe nanoparticles for effectively converting tar into gas. The reduction in tar content was proportional to the increase in H₂ content. The authors reported that the metal particles were evenly distributed and had a particle size of less than 20 nm.

In a different approach, Kang et al. [4] prepared a mesoporous FeNi/graphitic carbon nanocomposite catalyst. They used a cost-effective, non-pathogenic bacterium with naturally absorbed Fe–Ni ions as a carbon source, which was calcined to generate the final catalyst, BC-FeNi. The toluene conversion in continuous flow reaction over the proposed catalyst BC-FeNi was above 82.6% at 700 °C and 95.8% at 800 °C. The results were higher than those obtained using activated carbon-FeNi.

As reviewed by Guo et al. [1], nanocatalysts demonstrate high efficiency in tar removal. However, further efforts are still necessary for the commercial application of nanocatalysts in biomass thermochemical conversion, particularly in addressing the challenges related to the size of metals at the nanoscale, optimizing the structure of supports to enhance particle uniformity and dispersion, issues in operating on a larger scale, and developing low-cost regeneration routes for metals.

4.3.4. Physical and Mechanical Methods for Tar Removal

Physical/mechanical methods consist of separating tar from the hot gas (hot gas cleaning or dry method) or after cooling the gas down (wet gas cleaning). Hot gas cleaning comprises devices such as cyclones, rotating particle separators (RPS), electrostatic precipitators (ESP), filters (bag/baffle/ceramic/fabric/tube/sand bed), and adsorption. Cold gas cleaning is primarily based on wet scrubbers such as spray towers, cyclone spray, packed bed scrubbers (wash tower), impingement and venturi scrubbers, wet ESP, OLGA, and wet cyclones [167].

Wet scrubbing is a mature technique that has been widely explored and demonstrates good performance in removing particles and condensable tar from producer gas on a lab or industrial scale. The process consists of cooling the gas to temperatures below 100 °C followed by tar condensation and capture through droplets of scrubbing liquid. The phenomenon of particle impaction, absorption, and diffusion drives the mechanism [168]. Scrubbers are found in several designs, costs, and performances. The water spray tower is the simplest scrubber structure. Packed bed scrubbers involve a layered packing material inside the chamber to provide a surface area for gas/liquid contact. Venturi scrubbers are more expensive than other scrubbers but can remove up to 99% of particulate matter and 50% to 90% of tar [169]. The major problems of those wet methods are (i) the loss of energy because they require significant heating and cooling steps; (ii) the generation of liquid for disposal that must be further treated and/or recycled to avoid environmental impacts; and (iii) the tar potential chemical energy retained in the waste stream. The emulsion of tar in water is difficult to separate. The process uses cheap and easy-to-maintain equipment but becomes challenging in terms of scaling up and operation costs [168].

Water is the most used working fluid in wet scrubbers, but many organic compounds in tar have low solubility in water. Organic liquids or oil absorbents are considered suitable because they offer better performance in tar adsorption. Oil's lipophilicity characteristics help dissolve the non-polar hydrocarbon tar compounds [170]. Surjosatyo et al. [79] employed a venturi scrubber using acetone as a scrubbing liquid and compared its efficiency in tar removal with secondary thermal cracking. The optimum efficiency in tar collection of the venturi scrubber was 89%. Through secondary thermal tar cracking, the maximum efficiency was around 74%. Unyaphan et al. [171] proposed the use of emulsified oil absorbent (mixture of oil and water) to maximize tar removal for both polar and non-polar components. After cleaning the scrubber, emulsified oil (with 7.5% by volume of water) could remove 89.3% of tar. Pure canola oil only removed 75.0%.

To reduce the cost of using vegetable oil or rapeseed oil methyl ester (RMW), commercial absorbents and the use of waste cooking oil have been suggested. Lotfi et al. [80] used waste cooking oil as scrubber media in a bench-scale wet-packed bed scrubber. The removal of toluene and naphthalene was reported to be 70% and 90%, respectively. Sultan et al. [170] installed a wet scrubber system to eliminate tar using water or waste vegetable oil as scrubbing media. The overall tar removal efficiency was in the range of 48.45–82.16%, wherein vegetable oil provided the highest efficiency. Gabriele Calì et al. [172] designed a configuration of an updraft gasifier equipped with a wet scrubber for syngas clean-up. They integrated a wastewater treatment system to minimize water consumption and sludge disposal. The wastewater treatment, including an oil skimmer and an activated carbon adsorption filter, provided a 60% reduction in wastewater disposal.

OLGA is an oil-based gas washer developed by the Energy Research Centre of The Netherlands (ECN), considered one of the most efficient techniques for tar removal. It combines oil scrubbing columns with a stripper for oil regeneration. OLGA provides deep tar removal (up to 97%) without generating a wastewater emulsion of tar and water. The separated tar can be used to meet the heat demand of the gasification process [8].

The cleaning process in hot gas is performed at temperatures higher than 300 °C. This prevents heating loss since it does not require cooling and reheating of gas. Cyclones, RPS, and filter devices are predominantly used to remove particulate matter, which comprises solid material made up of ashes from inorganic compounds and unburned char. For effectively integrating tar and particulate abatement, conventional ceramic filters have been modified to be used as support catalysts [173]. A 15 wt% nickel-based ceramic filter was studied to clean syngas. It achieved a naphthalene conversion of 78% [173]. Tar content was reduced from more than 3000 mg/Nm³ to less than 450 mg/Nm³ from almond nut shell syngas [174].

Adsorption is a physicochemical technique widely employed for waste removal in many fields and has been observed as suitable for trapping lighter tar components. The process is based on the adhesion of tar molecules from the syngas on a solid material's surface (adsorbent) [175]. Activated carbon and biochar have been extensively studied for syngas adsorption. Due to the porous structures, they have high affinity and adsorption selectivity to hydrocarbon compounds [176]. Ravenni et al. [156] tested four wood-derived chars in the adsorption and decomposition of toluene and naphthalene. Due to the different heat treatments (gasification, activated-CO₂ pyrolysis, activated-H₂O pyrolysis, and pyrolysis), chars presented different surface areas and pore textures, resulting in different performances. They observed promising high adsorption capacity for activated carbon char at temperatures of 250 °C and below. A remarkable effect on naphthalene (up to 75.4 mg/g_{char}) was reported.

Heavy tar can block the porosity in the adsorbent, leading to poor capacity and quick saturation. Combining the adsorption technique with absorption or a wet scrubber is

recommended to overcome this problem. Heavy tar is mainly absorbed according to the dissolution and condensation principle, and light tar is adsorbed by the adsorbent connected downstream [177]. Singh et al. [176] proposed using biochar as a catalyst and absorbent for tar removal. As a result, 80% of tar was removed from syngas obtained by bio-oil mallee-wood steam gasification. Adsorption removed 60% of residual tar.

Compared to thermal and chemical methods, physical methods imply an increase in the complexity of gasification plants. Physical/mechanical methods have lower operating and maintenance costs than catalysts and thermal cracking; however, cleaning devices cannot reduce tar content to the required level for most applications. The cleaning process involves a series of devices to overcome limitations, and it must be combined with other methods. For example, to obtain the final tar concentration of 112 mg/Nm³ and 220 mg/Nm³, Awais et al. [164] submitted syngas through the cleaning system composed of a cyclone separator, wet scrubber, biomass filter, and auxiliary filter. The overall performance was 98.3% and 97.3% of tar removal from wood and corn cob gas, respectively.

4.3.5. Advanced Methods for Tar Removal

Another strategy that has been investigated for producing clean raw gas and as a cleaning technique for tar elimination is plasma gasification. In plasma gasification, an electrically ionized gas—gas that has split into free electrons and ions—produced in plasma torches at high temperatures—typically above 6000 °C—is heated by gasifiers. When it comes to the composition of the feedstock, the process is especially adaptable to high moisture biomass. It successfully breaks down organic matter into pure gas and inorganic minerals in molten slag free of tar or furans.

There are two types of plasma: thermal plasma, which has gas and electrons at the same temperature, and non-thermal plasma, which has electrons at a higher temperature. Commercial thermal plasma gasification is used to treat garbage, including hazardous or medical waste and municipal solid trash. Plasma technology is more likely to thermally fracture tar and produce very high-quality syngas that can be used to produce hydrogen and synthesize fine chemicals because of its extremely high temperatures [178]. Due to the high energy consumption needed to light torches and the potential for syngas impurities from nanoparticles to cause issues, plasma is subject to a number of technological and financial limitations [179].

Recent researchers have recognized non-thermal plasma (NTP) gasification for tar treatment. NTP operations occur at low temperatures (room temperature) of gases and ambient pressure, while electron temperature reaches a very high temperature. This non-equilibrium condition helps to overcome the constraints of high temperatures, requires significantly less energy, and allows the merging of catalysts to boost reactivity for plasma breakdown. Although the commercial scale is limited by process complexities, device lifetime, and cost, NTP catalytic is a promising novel technology for cleaning syngas [180]. Liu et al. [181] developed two gliding arc discharges (GAD) for biomass tar reforming. Magnetically accelerated GAD minimized the formation of undesirable liquid byproducts. The system reached a conversion of 71.4% for toluene and 87.5% for phenol. Tar models were converted into short-chain hydrocarbons, including C₂H₂.

The merging of catalysts and plasma can increase the catalytic activity and durability while improving the selectivity of the process [180]. Investigating the effect of NTP and NTP catalytic in naphthalene reforming, Cimerman et al. [182] showed that NTP catalytic has superior removal efficiency compared to catalytic or NTP treatment alone. A naphthalene removal efficiency of 88% was reached with a catalyst and 40% without. H Zhang et al. [183] performed plasma/catalytic tar reforming in rotating gliding arc discharge coupled with Ni-Cu bimetallic catalysts. The catalyst Ni₂Cu showed the anti-coke ability to operate

over 48 h with an average tar conversion of 90.3%. Ni-based catalysts supported on hydroxyapatite (Ni-HAP) and γ -Al₂O₃ (Ni- γ -Al₂O₃) were coupled with NTP to degrade biomass tar. Benzene conversion reached 92.31% at 400 °C [184]. Bin et al. [185] conducted a steam reforming of toluene in a dielectric barrier discharge plasma reactor combined with Ni/ γ -Al₂O₃ catalysts. At 450 °C, the process achieved a toluene conversion of 87.1% and a total gas yield of 72.6%. Table 4 summarizes some plasma findings reported in the literature.

Table 4. Reports on plasma technology for tar removal.

Plasma Technology	Objective of the Study	Findings	Ref.
Graded post-plasma	Thermal plasma biomass tar reforming using Ni-Fe supported with hydroxyapatite as catalyst	Benzene conversion of 84.8% at the discharge power of 100 W	[186]
Radio frequency plasma torch	Gasification of olive tree pruning in a rotary kiln reactor integrated with plasma torch	Integration of plasma decreased the benzene/toluene content from 8.3 g/Nm ³ to 2.2 g/Nm ³	[187]
Dielectric barrier discharge	Decomposition of toluene using 30 mm and 15 mm in length external electrodes at ambient temperature	Toluene decomposition >97% for 40 W of input power	[188]
Dielectric barrier discharge	Steam reforming of toluene in plasma reactor combined with Ni/ γ -Al ₂ O ₃ catalysts	Toluene conversion of 87.1%, total gas yield of 72.6%, and an energy efficiency of 18.2 g/kWh at 450 °C	[185]
Direct current thermal arc plasma	Wood pellet gasification in plasma torch using vapor as a main gasifying agent and a plasma-forming gas	Producer gas composed of H ₂ (43.86 vol.%) and CO (30.93 vol.%), H ₂ /CO ratio of 1.42 and LHV of 10.23 MJ/Nm ³ The tar content was in the range of 9.937 to 13.81 g/Nm ³	[161]
Dielectric barrier discharge	Catalytic gasification of rice hull by plasma over potassium catalyst	Gasification efficiency of 96.51% in the atmosphere of CO ₂ No tar was formed during gasification	[189]
Dielectric barrier discharge	Cracking of the toluene into C1–C6 hydrocarbons	Toluene removal efficiency of 92.8–98.4% using 10–40 W input power	[190]
Gliding arc discharge	Plasma reforming of biomass gasification tar coupled with honeycomb catalysts	Toluene and naphthalene conversion of 86.3% and 75.5% using honeycomb material coated with Ni/ γ -Al ₂ O ₃	[191]

5. Tar Removal in the Path of Circular Economy Approach

Fundamentally, lignocellulosic biomass gasification is a green technology that supports a sustainable circular economy by converting residues into energy and products. However, biorefineries must ensure that the cycle is maintained at every step of the process and throughout the entire biofuels production chain. There is increasing pressure on waste-to-energy systems to recover and valorize their residues and streams to achieve optimal carbon neutrality while ensuring economic feasibility for scaling up facilities. A greater emphasis has been placed on upcycling biomass-derived wastes, low-value materials and byproducts from other industries, and recovering materials from products at the end of their life to optimize the thermoconversion process. The reintegration of byproducts as a cost-effective source of catalysts is viewed as a promising strategy for enhancing the closed-loop biorefineries approach for syngas with a low carbon footprint.

5.1. Biochars

Biochar, the carbon-rich solid derived from gasification, pyrolysis, hydrothermal carbonization, or torrefaction, is a multipurpose material that can enhance soil properties, pollutant capture, and CO₂ storage. Physical properties such as porosity and surface area make this thermochemical product attractive, so it has recently become a research hotspot for replacing common catalysts in thermal processes [192]. This is a great pathway to pushing the economy to a carbon-neutral balance (Figure 6).

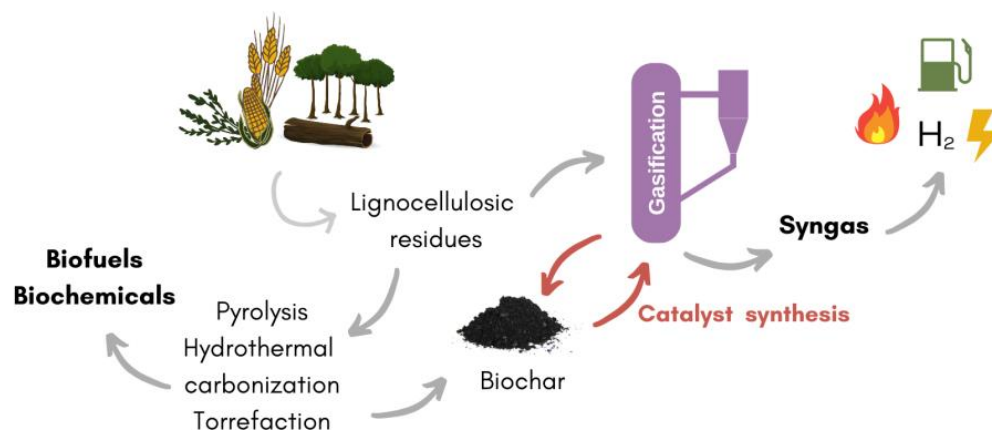


Figure 6. Reintegration of biochar for catalytic gasification and tar reforming.

Biomass-derived carbon materials are popular as heterogeneous catalysts because of their low cost, low toxicity, excellent thermal behavior, and broad applications. They are used as catalysts or support for loading micro/nano metal particles. Compared to other catalysts, biochar offers a simple synthesis route, strong sulfur and chlorine resistance, and easy catalyst regeneration after its deactivation [125].

The catalytic reactivity of char is attributed to four aspects: porous structure (micropores, <2 nm, mesopores, 2–50 nm, macropores, >50 nm), surface area, functional groups on the char surface, and the content of alkaline and AAEM oxides. The functional groups and oxides in the char surface and internal structure have selective catalytic effects and facilitate the ring-opening reaction of aromatic hydrocarbons [193]. The overall chemical structure and functionality of the resulting biochars depend on the types of biomass used and the production methods employed. Consequently, the efficiency, quality, and subsequent biochar application strategy depend on the quality of the raw material, production technology, and method conditions.

Muzyla et al. [194] compared the biochars obtained from the pyrolysis of wheat straw under various processing conditions. They found that pyrolysis temperature, grain size, residence time, and heating rate influence the volume and distribution of total pores, as well as the average pore diameter. At 500 °C, the biochars exhibited an underdeveloped surface area of 50–100 m²/g, whereas at 700 °C, the specific surface area reached nearly 400 m²/g. Additionally, a different pore distribution was observed. Although both biochars mainly contained micropores, higher temperatures resulted in a greater proportion of this fraction.

Biochars can be physically and chemically tailored using heat, acid/base processes, doping, and impregnation into various shapes, sizes, and forms to enhance productivity and selectivity. The efficiency of biochar in tar removal has been investigated, and several studies have reported on the effectiveness of different biochars while proposing methods for enhancing their properties and performance.

Dieguez-Alonso et al. [195] studied the impact of biochar modification on the adsorption and cracking of naphthalene as a model compound for tar. The authors produced biochar from beech wood chips by varying biomass pretreatment (H₂O washed, doped with

FeSO₄ or KCl), temperature (500–750 °C), and atmosphere (N₂, CO₂, H₂O, and CO). The highest adsorption capacity was achieved with biochar doped with FeSO₄ under a reactive atmosphere. For tar cracking experiments conducted at 850 °C, initial conversions of 100% were achieved for BW-700 °C, and 80% for BW-Fe-700 °C and BW-K-700 °C. However, after one hour, deactivation was observed, reaching 20% conversion. The biochars produced under a reactive atmosphere with untreated biomass and H₂O-washed biomass demonstrated the longest deactivation times. K-doping enhanced the adsorption capacity of biochars but negatively affected their performance as catalysts for naphthalene conversion.

In the work by Mbeugang et al. [196], biochar and a biochar-supported catalyst impregnated with Fe(NO₃)₃ and Ni(NO₃)₂ were prepared in N₂ or CO₂ atmospheres. During the gasification of pine sawdust, the pure biochar showed an effect only at 750 °C, where the H₂ content doubled compared to the process without catalyst addition. The Ni-Fe/biochar bimetallic catalysts exhibited excellent catalytic performance at all testing temperatures. The tar yield decreased from 11.44 mg/g biomass to 3.3 mg/g biomass at 750 °C with the inclusion of Ni-Fe/biochar-CO₂. Regarding catalyst preparation, both catalysts displayed a porous structure with a very similar surface area (330 m²/g) but different pore distributions. The pore structure distribution of biochar produced in CO₂ proved to be more advantageous for absorbing volatile compounds and outperformed biochar generated in N₂ in terms of tar removal.

Bhandari et al. [197] synthesized biochar and ultrasonicated-activated carbon from switchgrass for fluidized bed and downdraft gasification. All catalysts effectively removed tar, achieving an efficiency of 69–92%. However, the surface area of biochar increased from 64 to 900 m²/g after ultrasonication activation, leading to higher toluene removal efficiency. Downdraft gasification (900 m²/g) outperformed fluidized bed gasification (200 m²/g), and the reduction in surface area after 4 h of operation was 88% for biochar and 25% for activated carbons, respectively.

Abdelaal et al. [198] evaluated the performance of biochars produced from an industrial gasifier, a pilot-scale gasifier, and a lab pyrolysis reactor regarding tar removal efficiency. A significant difference was observed in ash content, oxygen content, surface area, and pore volume among the three chars. The industrial char showed the highest surface area of 1454 m²/g and demonstrated the best efficiency in tar removal, followed by the pyrolysis (805 m²/g) and pilot-scale gasifier (398 m²/g).

Using a one-step impregnation method, Bai et al. [199] prepared char catalysts derived from furfural residue. The tar conversion efficiency of biochar impregnated with the Ni catalyst reached 94.70% at 800 °C, yielding gaseous products of 681.81 mL/g. After five cycles of stability tests, the catalyst maintained a tar conversion efficiency of 85.90% and a gaseous product yield of 515.61 mL/g.

Ali et al. [200] prepared bimetallic nanocatalysts using one-step pyrolysis with biochar derived from Chinese herb residues as precursors. The biochar catalysts that were tested included BN (pure biochar), K/BN (10 wt% K₂CO₃), Fe/BN (15% Fe₂O₃), and Fe-K/BN (10 wt% K₂CO₃ + 5 wt% Fe₂O₃). Water washing was conducted to eliminate impurities accumulated on the surface of the biochar, enhancing the BET surface area from 1.36 m²/g to 384.42 m²/g. Pure biochar achieved only a toluene conversion of 40%. However, Fe-K/BN reached 100% toluene conversion at 650 °C after steam activation.

Hu et al. [125] compared thermal cracking and thermal catalytic reforming using biochar. As the temperature increases, the conversion of tar into permanent gases rises sharply in both thermal and thermal catalytic processes, though with different profiles and yields. The activation energy for each gas fraction, particularly for H₂, CO, and CO₂, was significantly lower in catalytic reforming. The total tar conversion stood at 48.9% for thermal cracking and 79.1% for catalytic reforming. The differing reaction behavior and

kinetics were attributed to the tar conversion mechanism. According to the authors, in tar catalytic reforming, hydrogenation and ring-opening reactions predominated over the poly-condensation reactions that prevail in thermal cracking.

Fuentes–Cano et al. [166] investigated the technical viability of using non-activated biomass biochar for tar conversion. Syngas was produced from the gasification of wood pellets in steam-blown BFB at 750 °C, followed by thermal treatment in a secondary reactor with non-activated char particles. The test conducted at 875 °C resulted in an initial tar conversion above 70% and over 64% after 5 h. During the experiment, the conversion of the heaviest tar was above 80%. Feng et al. [201] prepared pyrolysis walnut sawdust biochar and modified it by steam activation at 800 °C. Tar removal of 91.9% was obtained over 30 min-activated biochar. A superior tar removal rate of >70% was maintained for up to 45 min of operation. In the catalytic removal of toluene, the Fe-loaded biochar catalysts could remove up to 90% of toluene after 90 min of operations [193]. The high rate of catalyst deactivation due to coke deposition on the catalyst surface and in the pore channels is a major barrier in biochar catalysts.

Guo et al. [178] used potassium ferrate (K_2FeO_4) to create a biochar-supported K–Fe bimetallic catalyst in a single step, utilizing peanut shells (PSC) as the precursor. Pure PSC demonstrated moderate catalytic performance in tar cracking, achieving a maximum tar conversion of 83.4% at 800 °C. In contrast, the PSC- K_2FeO_4 catalyst showed promise as an effective catalyst for low-temperature operations, where the tar conversion efficiency reached 87.0% at 600 °C and 94.9% at 900 °C. The stability and reusability of PSC- K_2FeO_4 were assessed, with observations indicating that tar conversion efficiency consistently remained above 91% for five cycles.

Biochar, whether serving as a catalyst or as a support for one, demonstrates highly promising performance in the catalytic gasification of various biomass types. Biochar-based catalysts have been designed and prepared to enhance catalytic activity related to tar removal and to improve gas quality. However, further research in this field is necessary, as biochar displays a heterogeneous internal structure composed of inorganic and aromatic species, as well as a distinct porous structure that varies depending on the source and production method. This variability impacts catalytic activity and complicates the precise tailoring of catalysts for large-scale applications.

5.2. Alternative Materials

In the context of the circular economy, the reuse and recovery of materials from several industrial processes can also be embraced to reduce costs and optimize the thermochemical conversion of lignocellulosic biomass. In this sense, wastes from other sectors have been investigated for their influence on controlling tar during gasification.

Ajorloo et al. proposed the co-gasification of biomass and ethylene vinyl acetate (EVA) from end-of-life solar panels, which poses a growing challenge for management. They conducted gasification of pine sawdust and determined that the optimal conditions for maximizing gas components and minimizing tar production were $T = 790$ °C, $ER = 0.17$, and an EVA ratio of 25. The tar from pure biomass gasification primarily comprised oxygenates and nitrogenous compounds. Blending EVA with biomass was suitable for enhancing the quality of tar produced, as the amount of oxygenates in the tar decreases significantly, resulting in more non-oxygenated hydrocarbons.

Zhang et al. [179] proposed using $CaSO_4$ as a non-metallic gasification agent to replace metal oxides as solid-phase gasification agents. The authors suggest utilizing the hazardous solid waste phosphogypsum (PG), a byproduct of wet phosphoric acid production, which consists of 60–85% $CaSO_4$. Gasification experiments were conducted in a thermogravimetric analyzer, using rubber wood chips and solid waste corn straw. The PG, by releasing lattice

oxygen to oxidize biomass, proved to be more effective in tar cracking than pure CaSO_4 due to impurities such as Fe_2O_3 , SiO_2 , and Al_2O_3 .

Han et al. [202] developed a type of modified CaO sorbent that integrates carbide slag, a solid waste composed of $\text{Ca}(\text{OH})_2$ generated during the industrial production of polyvinyl chloride, with cement to enhance biomass calcium looping gasification (CLG). The cement-modified carbide slag demonstrated better performance than the natural sorbent, increasing H_2 concentration and yield while reducing the content of tar species. The tar yield from the steam gasification of corn cob was 0.341 g/g biomass. However, when 15% carbide slag and cement were added, the tar yield decreased by 14.1%. Additionally, the contents of light polycyclic aromatic hydrocarbons and heavy polycyclic aromatic hydrocarbons both decreased, while the phenols content increased in the presence of carbide slag, suggesting the catalytic decomposition of large tar species into smaller compounds.

Niu et al. [203] used Fe-ion-modified dolomite as a catalyst for the co-gasification of forestry waste (pine wood) and polypropylene waste (masks). A mixture with a pine wood-to-mask ratio of 1:2, along with a Fe loading of 8%, was found to achieve the maximum synergistic effect. Increasing the proportion of the masks in the feedstock enhances the volume fraction of H_2 , while pure masks reduce the quality of the gas. Regarding tar, the synergy between biomass decreased the phenol content, resulting in more complete tar cracking.

Sui et al. [204] studied biomass gasification in the presence of calcined cement to eliminate tar content and enhance fuel gas production. When gasifying wood chips with an additional cement loading of 0 to 9 wt%, the lead gas yield increased from 90.5 to 99.7%, while tar and char yields decreased from 6.92 g/ Nm^3 to 0.49 g/ Nm^3 . Thus, cement can be considered an abundant and inexpensive CaO -based additive with physical properties for tar cracking.

Parrillo et al. [205] investigated the removal efficiency of naphthalene, a model compound for tar, using two waste-derived material catalysts: red mud ($\text{RM}/\gamma\text{-Al}_2\text{O}_3$) and sewage sludge ($\text{SS}/\gamma\text{-Al}_2\text{O}_3$). The conversion efficiencies of naphthalene for the $\gamma\text{-Al}_2\text{O}_3$ -supported catalysts were consistent across all tests, remaining nearly constant and ranging between 60% and 80%. In comparison to pure iron-based catalysts, RM and SS effectively helped prevent sintering and the loss of catalytic elements during the process.

Abedin et al. [206] employed iron ore as a dual-function bed additive for the microwave-assisted gasification of corn stover mixed with waste plastics, upcycling it to generate syngas with greater hydrogen gas and lower tar yield. The strong synergistic effects and catalytically active plastic indicated the superiority of the system in terms of higher H_2 yield and lower tar formation. This reveals a potential for commercialization to improve biomass gasification while reducing plastic waste.

6. Conclusions and Prospects

The cleaning process for tar removal is a critical step in the further use of synthesis gas. The most important approaches for handling tar are classified into primary and secondary methods.

Secondary methods based on the use of physical and mechanical devices have lower efficiency in tar removal when used alone. High efficiencies can be achieved by adopting different equipment in a multi-stage cleaning process. The use of the candle catalyst filter has also been attractive for merging the removal of particulates and tar conversion. The development of new device designs and improvements in candle catalytic filters and wet scrubbers with alternative scrubber media will greatly contribute to optimizing syngas clean-up. In future research, the management of the generated hazardous waste has to be addressed.

Catalyst methods are preferable because they can promote complete tar destruction without toxic waste and improve gas yield and quality. Nickel catalysts have been widely applied in tar reforming because they intensify hydrogen production and removal efficiency. They are cost-efficient when compared to noble metals. Dolomite has shown remarkable results in tar reduction when enhanced by calcination or impregnated with metals (e.g., Ni, Co, Fe). Biochars have been exploited as catalysts and as a support because of their porous structure, high surface area, pore volume, and mineral content. When activated or loaded with metals on the surface, biochar performance in tar removal can be higher than 90%. However, considerable efforts are needed to improve biochar catalytic stability.

It was observed in this review that most studies are limited to lab-scale with tar-model compounds and controlled conditions; therefore, research based on real tar samples is recommended. In the field of catalytic reforming or gasification, some shortcomings must be addressed in future research for practical application in the real world: catalyst deactivation by coke deposition on the surface, deactivation of active sites by sulfur and chloride poisoning, development of a catalyst with high activity at lower temperatures to minimize energy consumption, modification of the catalyst to improve catalytic activity for all tar components, investigation of the combination of highly active catalyst and biochars or natural rocks to determine the optimal proportion to enhance performance and reduce costs and optimization of catalysts regeneration and tar recycling systems. Overall, the combination of primary and secondary methods is recommended to attain the desired syngas purity in terms of tar and particulate matter.

From another perspective, the combination of plasma technology and catalytic reforming has emerged as an interesting approach for tar conversions. Non-thermal plasma catalysts are still complex technologies, and more research is necessary to achieve cost viability and commercial scale. With further investigation, this technology seems to be decisive in biohydrogen and fine chemical products from gasification.

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