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Progress in Catalytic Gasification of Lignocellulose: Mechanisms, Catalysts, and Process Integration

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Abstract. Gasification of lignocellulosic biomass is now seen as a good way to produce renewable syngas, hydrogen, and valuable chemicals, while also helping with carbon neutrality and sustainable energy. However, problems like low efficiency, catalyst deactivation, tar formation, and limited process integration still stop it from being used on a large scale. This review gives an overview of recent progress in catalytic gasification of lignocellulosic biomass from the angles of reaction mechanisms, making catalysts, process strengthening, and combining systems. First, basic routes of tar cracking and methane reforming are summarized to show their roles in syngas upgrading and hydrogen production. Then, catalytic performance, deactivation behavior, and modification methods for natural minerals, alkali and alkaline earth metals, nickel catalysts, two-metal systems, and new support materials are discussed. Recent progress in process optimization, such as biomass chemical looping gasification, co-gasifying coal and biomass, microwave-assisted gasification, and adjusting operating conditions, is reviewed, with focus on improving syngas quality, carbon conversion, and energy efficiency. Moreover, advances in combined gasification systems that make heat and electricity together, hydrogen, and chemicals are looked at, noting their potential for being carbon-negative and producing several products. Finally, remaining technical problems about catalyst stability, process cost, feedstock variability, and industrial scale-up are pointed out, and future study on better catalyst design, linking processes, digital optimization, and combined energy systems is suggested. This review offers a systematic reference for developing and commercializing high-efficiency catalytic biomass gasification technologies.

Keywords: lignocellulosic biomass gasification, catalytic gasification, syngas production, process intensification, system integration

1. Introduction

The accelerating depletion of fossil resources, increasing greenhouse gas emissions, and the urgent need to achieve global carbon neutrality have stimulated extensive interest in renewable and sustainable energy technologies [1-4]. Among renewable resources, lignocellulosic biomass has emerged as a promising carbon-neutral feedstock because of its abundance, renewability, wide distribution, and ability to convert solar energy into chemical energy through photosynthesis [1, 5-7].

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Lignocellulosic feedstocks, unlike first-generation bioresources, do not compete with food production. They include agricultural residues, forestry wastes, energy crops, and industrial organic wastes, and this makes them attractive for sustainable energy use [3, 6, 8]. Among biomass conversion technologies, gasification is seen as an efficient thermochemical way to make clean fuels and platform chemicals [1, 2, 4, 9-11]. During gasification, the lignocellulosic biomass goes through pyrolysis, oxidation, reduction, and reforming reactions, and a synthesis gas (syngas) is formed that mainly has H_2, CO, CH_4, and CO_2 [11-21]. This syngas can be used for heat and power generation or upgraded to hydrogen, methanol, ammonia, Fischer-Tropsch fuels, and synthetic natural gas [1, 4, 21-33]. Compared with direct burning, biomass gasification usually gives higher energy efficiency, more product flexibility, and lower pollutant emissions [7, 14, 19, 34-37].

Even with these good points, biomass gasification still has technical and economic problems [2, 3, 38]. In thermal conversion, significant tar, char, and condensable hydrocarbons are produced, and they foul equipment, deactivate catalysts, make operation unstable, and reduce syngas quality [5, 39, 40]. Catalyst sintering, carbon deposition, sulfur poisoning, feedstock variability, and unfavorable economics further stop industrial application [9, 10]. Tar formation remains a big bottleneck as it cuts carbon conversion efficiency and harms downstream upgrading [1, 8, 26, 32, 41-43].

To solve these problems, researchers have worked on catalyst development and reaction engineering. Natural minerals like dolomite, limestone, and olivine have been widely studied because they are cheap and good at tar reforming [9, 13, 30, 44-48]. Akali and alkaline earth metals (AAEMs), especially K- and Ca-based catalysts, can boost gasification reactivity and hydrogen production [19, 49-53]. Nickel catalysts are the most studied because of their high activity for tar cracking, methane reforming, and syngas upgrading [18, 24, 54-56]. Recent progress on two-metal catalysts, creating oxygen vacancies, catalysts on biochar, and mixed supports has improved catalyst stability [12, 21, 28, 33, 35, 57-60].

Big steps have also been made in process intensification and system integration. Technologies like biomass chemical looping gasification (BCLG), microwave-assisted gasification, sorption-enhanced gasification, and coal-biomass co-gasification have raised carbon conversion, hydrogen yield, syngas quality, and energy efficiency [13, 18, 19, 25, 30, 31, 33, 34, 36, 57, 61-69]. BCLG is especially noted because it can produce nitrogen-free syngas and help capture carbon at the same time [57, 62, 70, 71]. Also, biomass gasification has been put together with hydrogen production, methanol making, ammonia production, fuel cells, and Power-to-X systems, and this widens its use in low-carbon energy setups [18, 19, 71-76].

Therefore, this review provides an overview of recent progress in catalytic gasification of lignocellulosic biomass. It talks about how tar cracking and methane reforming work, and then looks at catalyst types, their performance, and how they deactivate. Recent advances in process optimization, reaction strengthening, and system integration are summarized, and future directions on catalyst design, process cost, digital optimization, and making the process bigger are pointed out.

2. Catalytic gasification technology

2.1. Reaction mechanisms and pathways

Catalytic gasification of plant biomass has many reactions that are linked together, and these reactions decide the carbon conversion, the makeup of syngas, and how much tar is formed. The main steps include biomass pyrolysis, char conversion, and the catalytic breaking of tar and light hydrocarbons like methane. Tar cracking and reforming are especially important, because they raise the production of hydrogen and carbon monoxide, and they also cut down unwanted by-products. To

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design good catalysts and to improve the process, it is needed to understand these reaction mechanisms. This section gives a summary of the basic reaction pathways, and it looks closely at tar conversion and methane reforming, because these are the main ways to upgrade syngas and to make the gasification process work better.

2.1.1. Mechanisms of catalytic tar cracking

Tar is a complicated mix of organic hydrocarbons that are heavier than benzene, and it forms unavoidably during biomass gasification, with lignin as its main source material [5]. When tar forms, it causes carbon to deposit and catalysts to lose activity [6], it also lowers the quality of syngas, blocks pipes downstream, and damages equipment [5]. Tar can also stop microbes from working well, and this slows down the development of technologies that combine thermochemical and microbial ways to use biomass [8]. So catalytic cracking of tar is necessary, and the way it works can be broken into four steps: adsorption, activation, reaction, and desorption.

The process starts with adsorption, where tar molecules meet the catalyst surface. Catalysts that have very large surface areas, for example, the carbon-supported NiFe-$NiFe_2O_4$ catalyst made by Zhang et al. [63] (with 481.95 m^2/g), offer many small pores that physically grab tar molecules. At the same time, the ring-shaped parts in tar (aromatic rings) and groups that have oxygen stick onto active sites through chemical adsorption. The aromatic rings make π-complexes with metal sites like nickel [29], while basic sites on CaO attract acidic oxygenated compounds, such as phenols and carboxylic acids [40].

In the activation step, bonds in tar molecules are broken at active centers, and reactive middle products are formed. Catalysts that are based on nickel are good at cutting C-C, C-H, and C-O bonds, and so they activate the tar [6]. CaO catalysts help take away oxygen by breaking C-O bonds [40]. The carbon-supported NiFe-$NiFe_2O_4$ catalyst can also open the rings of large polycyclic aromatic hydrocarbons, and this makes the activation of complicated tar types faster [63].

In the reaction stage, the activated middle products are changed into small-molecule gases and light hydrocarbons. Then, depending on the gas atmosphere around them, these products go through reforming with steam, reforming with carbon dioxide, or the water-gas shift reaction (WGSR), and finally a hydrogen-rich syngas is made that contains H_2O, CO, and CO_2.

Table 1. Different reaction stages proposed by former scholars

Reaction	References
$C_nH_m \rightarrow C_n + H_2 + Coke$	[29]
$C_nH_m + nH_2O \rightarrow CO + H_2$	[77]
$CO + H_2O \rightarrow CO_2 + H_2$	[78]
$C_nH_m + H_2 \rightarrow CH_4 + C_nH_m$	[29]
$C_nH_m + CO_2 \rightarrow CO + H_2$	[29]
$CH_4 + H_2O \rightarrow CO + 3H_2$	[77]

The process ends with the desorption stage, where small-molecule products such as H_2, CO, and light hydrocarbons come off the catalyst surface and enter the gas phase. Timely and efficient desorption acts as a critical bottleneck, because it frees up blocked active sites and stops the catalyst from getting clogged. Higher temperatures, for example 800 °C, make gases like H_2 and CO desorb much faster, and in this way blockage of active centers is avoided [79].

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2.1.2. Reaction pathways of methane reforming

Besides tar cracking, making light hydrocarbons like methane into more useful gases is also a key part of improving syngas quality, and it is mainly done through reforming reactions. Methane reforming is an important technology for upgrading syngas in catalytic biomass gasification, and it aims to turn CH_4 into H_2 and CO, which raises the heating value and the use efficiency of the syngas that is produced. This upgrading process can be grouped into three main catalytic paths, namely steam methane reforming (SMR), dry methane reforming (DRM), and partial oxidation of methane. SMR is a mature technology for making hydrogen on a big scale, and its main reaction is shown as follows [57]:

$$CH_4\,(g)+H_2O\,(g)\leftrightharpoons CO\,(g)+3H_2\,(g),\ \Delta H=+206\ kJ/mol \tag{1}$$

SMR is strongly endothermic, so it strictly needs high temperatures to run well. In this path, the main job of advanced catalysts is to lower the activation energy for breaking C-H bonds, and by doing that they speed up the whole reaction rate [57]. When SMR is combined with carbon capture and storage methods, it offers a promising way for low-carbon hydrogen production, and this draws a lot of attention in industry [80].

DRM proceeds via another highly endothermic path under catalytic conditions [81]:

$$CH_4+CO_2\rightleftharpoons CO+H_2,\ \Delta H=+246\ kJ/mol \tag{2}$$

DRM also demands high reaction temperatures [82]. In DRM, catalysts are needed to help CO_2 break apart and to oxidize the CH_x intermediates. Nickel-based catalysts are widely studied because of their high activity and low cost, but serious carbon deposition often causes them to deactivate quickly [82]. So, developing catalysts that can resist coke remains very important for industrial uses of DRM. For example, the Ni-Ce/W-Zr catalyst made by Al-Fatesh et al. Uses a dynamic oxygen-vacancy supply system, and this system is first provided by the CeO_2-ZrO_2 solid solution and later kept going by the $Ce_2(WO_4)_3$ phase, and it continuously stops coke from forming during a 100-hour DRM test [12]. Also, when Ni nanoparticles are confined in a structure, for instance by cross-linked CeO_2 nanosheets, metal sintering can be stopped and carbon deposition can be cut down [82].

Partial oxidation of methane occurs through a mild exothermic pathway [81]:

$$CH_4\,(g)+\tfrac{1}{2}\,O_2\,(g)\rightleftharpoons CO\,(g)+2H_2\,(g),\ \Delta H=-149\ kJ/mol \tag{3}$$

Partial oxidation of methane can be operated at moderate temperatures (700-850 ℃). This reaction naturally gives syngas with an H_2/CO ratio near two, and this ratio is very desirable for later Fischer-Tropsch or methanol synthesis [81]. In this reaction, advanced catalysts offer active sites that adsorb and activate CH_4, and they break the C-H bonds through a step-by-step dehydrogenation process [83]. During this process, Ni-based or perovskite-type catalysts can make good use of their surface lattice oxygen to directly oxidize methane into CO and H_2 [81]. A key point is that the feed ratio of CH_4 to O_2 directly decides what products are made. When the ratio is below 0.5, the reaction tends to go through complete burning, mainly giving CO_2 and H_2O, but when this ratio is raised above 0.5, the reaction path moves toward partial oxidation, and then the generation of the target products, that is H_2 and CO, is greatly increased [81].

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2.2. Catalyst types and characteristics

Based on the fundamental reaction mechanisms, the selection of catalysts is very important for making gasification work better, for improving syngas quality, and for turning tar into something useful. This part reviews the main catalyst types—natural minerals, alkali and alkaline earth metals, nickel-based and two-metal catalysts, and improved support materials—and it focuses on how they perform, what problems they have with deactivation, and how they are changed to work better. It gives a clear summary of current progress and where research is going.

2.2.1. Mineral catalysts: catalytic performance and limitations of natural mineral materials

Catalysts made from natural minerals get used a lot, and this is because they are easy to find, cheap, and easy to get. They help a great deal with tar cracking and reforming during gasification, and so the amount of tar in the syngas is lowered [84]. Common mineral catalysts are dolomite ($CaMg(CO_3)_2$) [44], olivine ($(Mg, Fe)_2SiO_4$) [47], limestone (CaO) [84], trona ($Na_3(CO_3)(HCO_3)\cdot 2H_2O$), and borax ($Na_2B_4O_7\cdot 10H_2O$) [49].

Dolomite that has been heated (calcined) contains CaO and MgO phases. These phases can take in CO_2 and push the WGSR toward making H_2 [9]. The same oxides also give active sites for tar cracking and reforming [45]. For example, when calcined dolomite is added to a NiO/Pac-SD-1 catalyst system, the maximum H_2 concentration goes up from 51.14 vol to 62.54 vol, and the H_2 yield rises from 710.56 to 1343.52 mL per gram of biomass [13]. In pine gasification, using dolomite can bring the leftover tar content down to 1.87 $g/N\cdot m^3$ [10]. However, natural dolomite still has troubles with carbon build-up, mechanical wear, and its pores getting blocked by tar [45]. To make it work better, researchers have put forward the idea of adding promoters (Pt, Co, Fe, or La_2O_3) and using improved pretreatment ways, so that stability goes up and deactivation goes down [45, 46].

Olivine resists wear better than dolomite, so it is more suited for fluidized beds that have to run for a long time [44]. As a magnesium-iron silicate mineral, olivine holds active Fe and Mg parts that help make hydrogen [48]. When it is heated (calcined), the iron parts move to the surface and turn into active iron oxides, and this makes steam reforming and tar cracking better [47]. Using calcined Ni/olivine in two-stage gasification can push the H_2 yield up to 30.3 [48], and it also raises the H_2 content by 2.6-4.6 vol% in circulating fluidized beds [44]. In fluidized bed gasification with air, Ni/olivine lowers the tar yield from 6.8 $g/N\cdot m^3$ to 1.3 $g/N\cdot m^3$, and the tar capture efficiency goes up from 86.2% to 95.1% [47]. Still, natural olivine has its limits, like low reactivity by itself [30], high cost for grinding [47], and coke formation. Current ways to make it better are focused on heating (calcination), making the surface area bigger, and adding metals. For instance, CuO/NiO put on olivine uses the ability of Cu to let go of oxygen and the tar-cracking activity of Ni, and by doing this the gasification performance is made even better [30].

2.2.2. AAEM catalysts: broad availability and challenges of ashing and corrosion

AAEM catalysts that are often used mainly include materials based on potassium, sodium, calcium, and magnesium [85]. These catalysts come from plenty of natural minerals and cheap industrial wastes, and so they are easy to get and cost little. Biomass itself also contains many AAEM species naturally, and this gives several ways to obtain the catalysts [86]. In general, alkali metals show stronger catalytic activity than alkaline earth metals, with potassium-based catalysts giving the highest efficiency.

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K_2CO_3 is one of the most widely applied and active potassium-based catalysts, and its high activity comes from the fact that potassium can react with carbon to form intercalated compounds. This widens the space between carbon layers and makes the C-C bonds weaker, and as a result gasification and CO generation are sped up [53]. At the same time, potassium makes many active sites inside the material, and it helps release oxygen and promotes its absorption by carbon, and this greatly speeds up the Boudouard reaction;

$$C + CO_2 \rightleftharpoons 2CO \tag{4}$$

In the devolatilization step, K_2CO_3 helps biomass depolymerize and makes lignin and hemicellulose decompose faster, which raises the production of CH_4 and CO_2 [87]. When steam is present at high temperature, K_2CO_3 can also take part in homogeneous Catalysis as hydroxide species, and it helps open aromatic rings, break side chains, crack tar, reform volatiles, and drive the WGSR [50]. Because potassium can move and spread more easily, it usually works better as a catalyst than alkaline earth metals such as calcium [53]. In corn stalk gasification, K_2CO_3 pushes the CO content from 16.02 vol% to 19.76 vol%, lifts the heating value of syngas from 5.38 MJ/N·m^3 to 6.43 MJ/N·m^3, and cuts the tar amount from 4.63 g/N·m^3 to 3.77 g/N·m^3 [87]. During lignin gasification with steam, it raises the H_2 yield from 12.80 mmol/g to 64.56 mmol/g and the carbon conversion from 35.14% to 72.03% [50]. However, potassium-based catalysts cannot stop H_2S and SO_2 emissions very well, and they tend to turn into vapor at high temperatures. To deal with these problems, K_2CO_3 is often used together with CaO. CaO captures SO_2 and forms $CaSO_4$, while $K_2Ca(CO_3)_2$ forms when the K:Ca ratio is set in the right range, and this creates a combined effect that makes the gasification reaction work even better [53, 87].

CaO is another catalyst that is widely used, and it is a calcium-based material. Its outstanding properties come from three aspects. First, CO_2 can be captured in situ by CaO, which turns it into $CaCO_3$. This shifts the WGSR equilibrium toward making more H_2, and so hydrogen-rich syngas yields are greatly increased while CO_2 is reduced [14, 88]. Second, the surface of CaO has many basic sites. These sites can upset the electron arrangement of the ring structures in tar polycyclic aromatic hydrocarbons, and as a result the C-H, C-C, and aromatic C=C bonds are cleaved more easily, which helps tar cracking and reforming into small molecules [88]. Furthermore, the carbonation reaction ($CaO + CO_2 \rightarrow CaCO_3$) is a strongly heat-releasing step ($\Delta H \approx$ -178 kJ/mol). It gives off a lot of thermal energy that can make up for the heat needed by endothermic reactions, like the breakdown of biomass and the Boudouard reaction. This creates a "thermal compensation" or "self-heating" effect, and it makes gasification efficient with less heating from outside [15]. Many studies have shown that CaO works well for raising H_2 production. Panichkittikul reported that when bagasse steam gasification was combined with CaO sorption, hydrogen was made with a purity of 99.95%, and with CaO-assisted supercritical water gasification, the H_2 purity went up to 99.99% [65]. Zhang et al. Found that with a CaO/cellulose mass ratio of 5, the H_2 yield rose to around 4 mmol/g, which was nearly eight times higher than the yield without catalyst, and the H_2/CO ratio remained about five times greater than that from normal CO_2 gasification after external power was removed [15]. Similarly, under the combined action of CaO and steam (steam-to-biomass ratio = 7.5, CaO-to-biomass ratio = 1), the H_2 yield reached 399.90 mL/g, about three times that of the untreated sample [88]. In rice straw gasification, CaO brought down the CO_2 concentration from 27.78 vol% to 5.37 vol% and raised the H_2 concentration from 33.32 vol% to 45.31 vol%, and so syngas quality was much improved [14].

Even though CaO captures CO_2 well and acts as a good catalyst, it still faces some problems. First, CaO is easily sintered, and its pores get blocked in high-temperature conditions. Pure CaO has

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a low specific surface area of only about 15.25 m^2/g, and the used particles often stick into irregular clumps because of crystal fusion, which causes a serious drop in porosity [14]. Second, heavy carbon deposition is a main cause of CaO deactivation, as the coke formed covers the active sites and weakens catalytic stability [14]. In addition, the SiO_2 in biomass ash can react with CaO to form calcium silicate (Ca_2SiO_4), and this leads to deactivation by reaction and lowers the catalytic efficiency [16]. Finally, the CO_2 sorption ability of CaO-based catalysts tends to fall after many cycles of heating and CO_2 absorption, so the cycle stability still needs to be made better [14, 5]. To solve these bottlenecks, several modification methods have been put forward. Mixing CaO with biochar that has good pore structures can greatly increase the specific surface area (from 15.25 to 70.27 m^2/g), and this improves CaO dispersion and stops sintering [14]. Adding transition metals such as Ni can improve the breaking of C-C and C-H bonds, which raises catalytic activity and resistance to coking and also helps keep the porous frameworks [14]. Additionally, treating CaO with a ternary molten salt (Li_2CO_3-Na_2CO_3-K_2CO_3) forms a liquid salt layer at high temperatures, and this helps CaO spread out, prevents sintering, and stops the unwanted reaction between CaO and SiO_2 in ash, so that deactivation by reaction is avoided [16].

2.2.3. Ni-based and bimetallic catalysts: high activity and deactivation mechanisms

During the catalytic gasification of lignocellulosic biomass, monometallic Ni-based catalysts are widely used because of their high activity and low cost. Common systems include oxide-supported catalysts (Ni/Al_2O_3, Ni/MgO, Ni/TiO_2, and Ni/CeO_2) [24], mineral-supported catalysts (Ni/Dolomite and Ni/Olivine) [24, 89], zeolite-supported catalysts, and carbon-based supports such as biochar and activated carbon (AC) [24, 89]. Their excellent performance mainly arises from high Ni dispersion and small particle sizes, which promote the Boudouard, dry reforming, and WGSR reactions, accelerating tar conversion into CO, CO_2, H_2, and CH_4 [20, 90, 91]. For example, Ni/Al_2O_3 increases H_2 selectivity to 51.78% during banana pseudo stem steam gasification [90], while Ni-Ce/ZrO_2 achieves an H_2 yield of 12.9 mmol/g during supercritical water gasification of rapeseed straw [90]. However, carbon deposition, metal sintering, and acidity-induced side reactions often lead to catalyst deactivation. To address these issues, nanoscale synthesis methods, alkaline earth metal doping, and CO_2-assisted gasification have been proposed. For instance, Sr-modified catalysts reduce the coking rate to only 0.34 wt% [17].

To further improve activity and stability, bimetallic Ni-based catalysts such as Ni-Fe, Ni-Co, and Ni-Cu have been developed. Among them, Ni-Fe catalysts exhibit particularly strong performance because alloy formation enhances Ni dispersion, lowers reduction temperatures, and promotes water activation [54]. They also accelerate WGSR and provide mobile oxygen species that facilitate coke removal. Experimentally, a Ni-Fe-BC@CO_2 catalyst achieves an H_2 yield of 1452.72 mL/gbiomass and reduces tar yield from 52.22 mg/gbiomass to 3.3 mg/gbiomass during steam gasification [21]. A Ni-Fe/CeO_2/CNT catalyst maintains over 80% toluene conversion for 50 h at 700°C [54], while Ni-4%Fe/HZSM-5 increases both H_2 and CO yields and reduces carbon deposition from 800 to 230 mg/g_{cat} under CO_2 conditions [92].

Nevertheless, Ni-Fe catalysts still suffer from carbon deposition and thermal sintering under harsh operating conditions [54, 93]. To improve durability, composite supports such as CeO_2/CNT can enhance oxygen transfer, thermal conductivity, and metal dispersion [54]. Introducing promoters such as Ca can further suppress nanoparticle agglomeration and improve anti-sintering performance [93]. In addition, optimizing the reaction atmosphere through appropriate CO_2 addition and steam-to-carbon ratios can promote in-situ coke removal and extend catalyst lifetime.

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2.2.4. Novel carrier materials: performance optimization and stability enhancement

Conventional Ni-based catalysts have problems with deactivation and poor stability, and to solve these, researchers have made several advanced support materials, for example biochar-based carriers, perovskite oxides, changed natural minerals, and composite supports. Among these, biochar-supported catalysts have got much attention, because they are cheap, they come from waste, they are carbon-neutral, and their pore structures can be tuned. Typical systems are monometallic catalysts like Ni/Biochar, Fe/Biochar, and Ca/Biochar [58, 94] and bimetallic ones like Ni-Fe/Biochar, Ni-La/Biochar, and Ni-Ce/Biochar [59, 95]. The high activity is because they have large surface areas, well-developed small-pore structures, and strong links between the metal and the support, which help tar move, make the metal spread out well, stop sintering, and make steam activation better [59, 94, 96, 97]. Also, the biochar itself contains alkali and transition metals that can take part in the catalytic reactions [98].

For example, a Ni-La/Biochar catalyst with 10% La can give 93% tar conversion and 87% H_2 yield at just 400 °C [59]. Under gas conditions that mimic cement flue gas with a lot of CO_2s, a Ni/AC catalyst makes the total gas amount go up by 21 times, and the H_2 and CO selectivities can reach 95% [97]. Similarly, when a Fe-Ni/biochar catalyst is used under a microwave field of 488 W, tar model compounds can be fully broken down through the effect of micro-discharges [95].

Even with these good points, using them on a large scale still has serious deactivation problems. When the biochar support is put in high-temperature steam or industrial flue gas, it starts to gasify by itself, and then the pores collapse, the places where the metal is anchored are lost, the metal particles come together, and coke is deposited [94, 97]. Also, if a one-step way of making them is used, the volatiles from the biomass can form carbon layers that cover the active metal sites, which reduces the catalytic performance [99]. To deal with these troubles, some ways to change the support have been put forward. Doping with boron, for example, can form C-O-B bonds and B_2O_3 active sites, and this cuts down the loss of biochar. A Ni-3B/FRC catalyst shows only 7.7%-9.6% loss of support mass after five gasification cycles [96]. Adding metals that like oxygen (such as Ce, La, Fe) or metal carbonates (such as $CaCO_3$ and $MgCO_3$) can greatly increase the oxygen vacancies and basicity on the surface, and this speeds up the adsorption and breaking of water, so coke build-up is stopped [59, 100]. As for how they are prepared, a two-step strategy can be used: first the biomass is pyrolyzed to get biochar, and then the metal salts are soaked in, and this keeps the active sites from being wrapped again by volatiles, so the abilities for hydrogenation and reforming stay higher [99].

2.3. Process optimization and intensification

Catalyst performance alone cannot determine gasification outcomes. Reactor design, feedstock, energy input, and operating conditions are equally critical. This section discusses process coupling, feedstock synergy, novel energy fields, and operating parameter optimization, highlighting strategies that improve syngas yield, hydrogen production, and tar reduction while advancing toward scalable and efficient gasification systems.

2.3.1. Process coupling and feedstock synergy

Biomass gasification technology is moving from standalone processes to integrated systems, and these systems can make the heat use better, allow more types of feedstocks, and improve product quality. This trend mainly shows in two directions, which are process coupling, with BCLG as a typical example, and feedstock synergy, shown by coal-biomass co-gasification. BCLG uses a

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circulating oxygen carrier (OC) that moves between a fuel reactor and an air reactor, and in this way the gasification is split into two sub-reactions that are connected [101]. It provides oxygen through lattice oxygen, not by blowing air directly, so nitrogen is not brought in, and syngas without nitrogen is made, and expensive air separation units are not needed [61]. Besides, when the OC is oxidized in the air reactor, heat is given off, and this heat is carried back to the fuel reactor to drive the biomass pyrolysis and gasification, and a self-sustained heat balance is reached [101]. For instance:

$$4Fe_3O_4+O_2\rightarrow 6Fe_2O_3,\ \Delta H=\text{-}471.96\ MJ/kmol \tag{5}$$

At the same time, the OC also works as a catalyst, and it helps crack tar, and the tar yield is cut by about 2.5 times compared with using inert bed materials, and NH_3 and NO_x precursors are turned into N_2 [62]. The fuel reactor produces a stream with concentrated CO_2, and this is very suitable for carbon capture and storage, and when it is combined with biomass, a life-cycle carbon footprint of -14.58 kg-CO_2 per kg-H_2 can be reached [62]. Because of this, BCLG is seen as a promising negative-emission technology that has high efficiency and low pollutant emissions [62]. Research on OCs has moved from single metal oxides to two-metal carriers like $CuFe_2O_4$ and $CoFe_2O_4$, because these can make the catalytic performance better [101]. Results from DFT studies show that $CuFe_2O_4$ adsorbs CO more strongly and has higher surface reactivity than $CoFe_2O_4$ and Fe_3O_4 [101]. ReaxFF molecular dynamics (MD) simulations have also shown more about how cellulose, hemicellulose, and lignin gasify at temperatures from 1000 to 1500 K [101]. Also, a solar-assisted Solar-BCLG-GT-ORC system can get an energy efficiency of 31.18%, and this goes up to 34.96% after the ORC part is added, and this is much higher than the usual biomass burning power plants [71]. BCLG mainly works on making the process better for one kind of biomass, but feedstock synergy strategies use biomass and coal together, and they make good use of the fact that their compositions help each other, and this brings an overall improvement of gasification performance. When coal and biomass are gasified together, the AAEMs (K, Na, Ca, Mg) that are in biomass ash work as natural catalysts, and they lower the activation energy and raise carbon conversion [36, 95]. K and Fe show clear synergistic effects, they help pores to form and make the gasification more reactive [95]. The organic parts of biomass also help break down the structure of coal, and this further reduces the overall activation energy [36]. In addition, AAEMs make the carbon structure more disordered and change the properties of the ash, and as a result the ash fusion temperatures go down and slagging becomes less likely [66]. Raw biomass does not grind well and has low energy density, so torrefaction pretreatment can change it into a fuel that is like coal, with higher heating value and more carbon. When torrefied biomass is co-gasified with low-rank coal, the syngas made is almost free of tar, the H_2 and CO yields go up, and carbon conversion and cold gas efficiency get better than with pure coal gasification [67]. A staged co-gasification system has also been put forward, and this system splits the process into co-coking, co-gasification, and external combustion. In this system, biomass acts as a hydrogen donor and helps tar cracking, the cold gas efficiency is raised to 83.21%, and with carbon capture before combustion, a power generation efficiency of 40.03% and a negative carbon footprint of -308.1 kg CO_2 per MWh can be achieved [68]. Because co-gasification systems are complex, machine learning models, for example gradient boosting regression (GBR) together with genetic algorithms, have been used to optimize the process. These models can predict syngas yield, H_2/CO ratio, and heating value with good accuracy ($R^2 > 0.92$, MAE < 2.92), and they suggest that biomass blending ratios of 50%-70% are good, and steam gasification gives the best economic results [25].

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2.3.2. Novel energy fields and reaction intensification technologies

In lignocellulose gasification, the use of unconventional energy forms—light, electricity, magnetism, microwaves, and plasma—to replace or assist traditional heat sources is what is referred to as novel energy fields and reaction intensification technologies. These technologies make heat transfer, mass transfer, and catalytic reactions stronger through specific physical ways, and as a result, the conversion processes become more efficient and are aimed at specific targets. Among these, microwave technology is the most widely applied novel energy field method, and it has the highest level of technology readiness.

The main working principle of microwave-assisted biomass gasification is its unique way of heating the whole volume. In this mode, microwave energy goes into the biomass and produces heat from the inside, and this creates a temperature gradient that goes from inside to outside, which is different from ordinary heating by conduction [64]. This method allows fast and selective heating, and it can also lower the activation energy needed for chemical reactions [19]. Because lignocellulosic biomass does not absorb microwaves well, people often add materials like silicon carbide (SiC) and biochar (BC) as susceptors to help take in the energy [64, 102]. Under microwave irradiation, these materials create local hot spots, and sometimes microwave plasma, and these give energy levels that are much higher than the overall temperature of the reaction [31]. As a result, the heavy tar molecules are cracked efficiently, which reduces the tar content and changes BC and heavy parts into H_2, CO, and CH_4 [31, 64]. Microwave heating also pushes forward the Boudouard reaction and the water-gas reaction, and this further raises the yield of syngas and the production of hydrogen [31].

In recent years, big progress has been made in microwave-assisted biomass gasification on reactor design, the optimization of working conditions, the study of energy conversion, and the understanding of micro-scale mechanisms. At the reactor level, researchers have built microwave-assisted fluidized bed reactors (to gasify rice husk) and U-shaped quartz tube reactors (to gasify corn stalk), and with these they can monitor temperature, gas compositions, and heating value online in real time, which supplies key data to help make the process bigger [63, 64]. Careful studies on the operating conditions show that an optimal loading amount exists for the microwave-absorbing materials, for example 30 g for SiC and 10 wt% for BC. When the amount goes above this value, the gasification efficiency drops because the microwaves cannot go as deep and local overheating occurs [31]. By choosing the right amount of susceptor, the specific energy consumption can be cut by 39%, and under the best conditions, the process efficiency reaches 19.7% and the cold gas efficiency is 55.6% [102]. Early assessments of cost and environment show that for a small-scale microwave gasification system, the cost to make electricity is about 586 USD per kW, and for every 1 kWh of energy produced, the global warming potential is only 0.08 kg CO_2 equivalent, and this points to a promising future for using it [103]. Overall, microwave-assisted gasification is now moving from laboratory-scale tests to bigger pilot-scale expansion. By optimizing the susceptor material, controlling the temperature, and making the reactors better, people expect it to reach the goal of using lignocellulosic biomass for energy in an efficient and clean way.

2.3.3. Operating parameters

Even when novel energy fields can provide improved reaction environments, precise control of operating parameters is still very important for making syngas production better and for reducing tar. During catalytic gasification, different kinds of catalysts have different roles. AAEMs mainly help with the WGSR and capturing CO_2 where it is formed, and this increases the H_2 amount and also

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helps crack tar [51]. Natural minerals like dolomite and olivine are cheap catalysts, and they can raise the syngas yield and H_2 production through active sites and free iron phases [91]. Catalysts that contain nickel are highly active for breaking bonds and for steam methane reforming, and so they greatly increase the production of H_2 and CO [91]. Precious metal catalysts are very good at tar cracking and resist coking, but they cost too much, and this limits their use on a big scale [2, 38].

The type of gasifying agent and the operating conditions have a strong effect on syngas composition, heating value, and tar content. Air gasification is cheap, but the syngas produced has a low heating value (4-7 MJ/N·m^3) because nitrogen from the air dilutes it [1, 11]. Oxygen gasification can give syngas with higher quality (12-28 MJ/N·m^3) and less tar, but it needs costly systems to make oxygen. Steam gasification raises hydrogen production by speeding up the water-gas reaction, carbon gasification, and methane reforming, and the syngas has heating values of 10-18 MJ/N·m^3 [1, 37]. People generally recommend a steam-to-biomass ratio of 0.5 to 1.5, and the best hydrogen yield is got when this ratio is near 1. Gasification with CO_2 mainly speeds up the Boudouard reaction, and this makes CO concentration go up and H_2/CO ratio go down [9].

Temperature is one of the operating parameters that has a very big influence. When the temperature goes up, generally more H_2 and CO are formed because the WGSR and steam reforming are helped at high temperatures [3]. If the temperature is increased from 650 °C to 950 °C, H_2 and CO become the main gas products [3]. Higher temperatures also make CH_4 and CO_2 concentrations go down, because methane reforming and the Boudouard reaction are made faster [104]. In addition, the thermal cracking of tar is made much stronger, and tar amount drops a lot when the temperature rises from 800 °C to 900 °C [4, 104]. But if the temperature goes too high (above 900 °C), it may cause the catalyst to sinter, carbon to deposit, and the catalyst to be damaged by heat, and all this can deactivate the catalyst.

Pressure also affects gas composition and tar formation a lot. Raising the pressure helps hydrogenation and methanation reactions, and this increases the yields of CH_4 and CO_2 while cutting down the production of H_2 and CO [7, 60]. Higher pressure can make the initial reaction rate go up (R0.5 increases by 70%), but limits in gas discharge may lower the final carbon conversion [60]. Its effect on tar formation is more complex, because pressure can either help crack tar, or it can make polycyclic aromatic hydrocarbons like naphthalene build up because the residence times get longer [7]. The final result depends strongly on the catalyst type and the operating temperature.

2.4. System integration and application directions

Besides improving catalysts and processes, it is key to make gasification part of real energy and chemical production systems. This section looks at multi-product systems, at making syngas better for hydrogen and chemicals, and at overall approaches that boost energy efficiency, product worth, and carbon reduction, with their tech possibilities and industrial importance being pointed out.

2.4.1. Gasification-power polygeneration systems

Systems for biomass gasification and power that make many products have developed. They moved from simple electricity making to combined heat and power, multi-product setups, and integrated systems that include carbon capture and fuel cells. In a basic layout, the syngas gets burned to make heat for power. But energy use is still limited because the subsystems run independently. Gas engines fueled by syngas usually reach electrical efficiencies of 20 to 35 percent. However, combined heat and power systems with waste heat recovery can go above 60 percent and reach total efficiencies up to 90 percent.

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Besides electricity, syngas can also be used to make bioplastics, biofuels, and single-cell proteins. For instance, a system that gasifies coconut shells with supercritical water makes syngas, biochar, heat, and electricity all at once. Its energy efficiency is 60.9 percent. Similarly, a system that combines biomass gasification, ammonia production, and power reaches an overall efficiency of 58.6 percent. This is much higher than the usual ways of making ammonia from biomass.

At a more advanced level, biomass gasification can be integrated with the Allam cycle. This can capture nearly 100 percent of carbon emissions and give a net-negative carbon footprint of -0.7445 kg CO_2 per kWh. Also, flexible systems called "Biomass + Power-to-X/Power" join gasification with reversible solid oxide cells. These systems can switch between making fuels and producing power, based on conditions in the electricity market.

In China, commercial-scale deployment has already been reached. The Xinjinyuan Project in Heilongjiang is an energy-oriented model. It combines the production of power, heat, biochar fertilizer, tar, and hydrogen. The installed capacity is 40 MW, and it consumes about 400,000 tons of biomass every year. Another example is the Huajing Apricot Shell Gasification Project in Hebei. This project focuses on producing AC, and it also uses the syngas for power generation and waste heat recovery. This forms a high-added-value way to use biomass.

2.4.2. Syngas for hydrogen production and chemical syntheses

Besides making electricity, syngas can be upgraded to hydrogen and platform chemicals. This is done through combined purification and conversion steps. For hydrogen production, steam is usually chosen. It helps the WGSR and SMR reactions [106]. CaO can capture CO_2 right where it is produced. This raises the H_2 concentration to 80-90% [34]. Modern plants that turn biomass into hydrogen combine many steps. These steps include dust removal, desulfurization, dealkalization, WGSR, and pressure swing adsorption units. The pressure swing adsorption unit gives hydrogen purities of 99.9-99.999%. The recovery rates are 80-95% [41]. When carbon capture and storage technologies are added, the potential for net-negative carbon emissions is made larger [106].

Syngas can also be turned into methanol, DME, acetic acid, Fischer-Tropsch hydrocarbons, and synthetic natural gas [23, 32, 74]. To make green methanol, oxygen-steam gasification is generally used. Strict removal of tar and impurities is also needed [39]. Pressure swing adsorption, sorption-enhanced reforming, and SOEC are often used together to improve the syngas composition [107, 108]. To lower production costs, recent studies have looked at multi-product systems [109]. These systems combine solar energy with making methanol, DME, and acetic acid. They can achieve 67.43% exergy efficiency. They lower global warming potential by 39.19%. The payback period is shortened to 6.56 years [74].

In 2025, a demonstration project by CIMC Enric in Zhanjiang, China, became the world's first commercial-scale green methanol facility. It uses only biomass gasification. The plant makes 50,000 tons per year. It achieves product purity of 99.9%. Life-cycle emissions are reduced by over 85%. Production costs are cut by about 30% compared to routes that use renewable electricity to drive electrolysis. However, for use on a large scale, capital costs need to be lowered. Biomass supply chains must be secured. Catalyst durability also needs to be improved.

3. Conclusion and future perspectives

Catalytic gasification of lignocellulosic biomass is a promising pathway, and it can turn renewable carbon resources into hydrogen-rich syngas, sustainable fuels, and valuable chemicals. In this review, recent advances in reaction mechanisms, catalyst development, process intensification, and

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system integration have been summarized. The key routes that control syngas quality and hydrogen production are tar cracking and methane reforming, and a better understanding of the mechanisms has given useful directions for designing catalysts and processes.

Natural minerals, AAEM catalysts, nickel-based catalysts, and new biochar-supported catalysts have all shown big potential. They can raise carbon conversion, take away tar, and make hydrogen generation better. At the same time, process intensification technologies have been developed. These include chemical looping gasification, microwave-assisted gasification, sorption-enhanced gasification, and coal-biomass co-gasification. They have made syngas quality, energy efficiency, and carbon use much better. Also, combining gasification with CHP systems, carbon capture technologies, fuel cells, and Power-to-X setups has given biomass gasification a wider role in future low-carbon energy systems.

Even with these advances, there are still problems. The problems are about catalyst lifetime, differences in feedstocks, handling tar, and the economics of the process. These still block large-scale commercial use. Future work should aim at making catalysts that are tougher. It should also aim at making the understanding of mechanisms stronger, making process integration better, and speeding up tests at industrial scale. If catalyst engineering and system optimization keep moving forward, catalytic biomass gasification is expected to play a bigger and bigger part in sustainable hydrogen production, renewable chemical making, and the shift to carbon-neutral energy. Future research should therefore focus on the following points.

Design highly stable catalysts, and they need better resistance to coking, sintering, and poisoning.

Make catalyst-support structures that can do multiple jobs, and they should help tar cracking, methane reforming, and carbon capture at the same time.

Bring in AI, machine learning, and digital models to improve the process and control operations.

Push forward gasification systems that are carbon-negative, and this can be done by linking them with technologies that capture and use or store carbon.

Stablishing economically competitive and scalable gasification-based biorefineries capable of producing multiple energy carriers and chemicals.

Overall, catalytic lignocellulosic biomass gasification is evolving from a conventional thermochemical conversion technology into a highly integrated renewable energy platform. Continued advances in catalyst engineering, process intensification, and system integration are expected to accelerate its commercialization and strengthen its contribution to global carbon neutrality and sustainable energy development.

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