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Catalytic Reforming for Hydrogen Production from Biomass Gasification

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Abstract: ***Hydrogen, as a clean and efficient energy carrier, plays a pivotal role in the global transition toward carbon neutrality. Biomass gasification coupled with catalytic reforming offers a promising route for the sustainable production of hydrogen by valorizing abundant renewable biomass resources, enabling low-carbon and even carbon-negative hydrogen generation. This review systematically summarizes recent progress in biomass gasification-catalytic reforming for hydrogen production, with a particular focus on the catalytic performance, stability, and reaction mechanisms of nickel-based, iron-based, noble metal-based, alkali/alkaline earth metal-based, bifunctional, and biochar-based catalysts. The effects of key operating parameters, including gasification temperature, steam-to-biomass ratio, and catalyst loading, on hydrogen yield and syngas quality are critically discussed, and the optimal operating windows for process control are identified. In addition, the major deactivation pathways of catalysts are analyzed in depth, including the multistage formation of carbon deposits and their morphological classification, as well as the underlying causes of metal sintering and poisoning. Corresponding mitigation strategies, such as bimetallic synergy, core-shell confinement structures, promoter incorporation, and optimized regeneration methods, are also summarized. This review aims to provide a comprehensive overview of the current state of biomass gasification-reforming technologies for hydrogen production and to offer insights into future research directions.***

Keywords: Biomass gasification, Catalytic reforming, Hydrogen production, Catalyst systems.

1. Introduction

The continued consumption of fossil fuels and the environmental impacts associated with greenhouse gas emissions have intensified the global demand for clean and renewable energy sources. Hydrogen, owing to its high energy density of approximately 142 MJ/kg, water as the sole combustion product, and wide availability, is widely regarded as a key energy carrier for future low-carbon energy systems [1]. According to the International Energy Agency, global hydrogen demand reached approximately 95 million tons in 2022 and is projected to exceed 180 million tons by 2030 [2]. However, around 70% to 80% of hydrogen is still produced via natural gas steam reforming, which typically emits 8-10 kg of CO_2 per kilogram of H_2 generated. Consequently, the development of low-carbon or even carbon-negative hydrogen production pathways has become an urgent priority.

Biomass is the only renewable carbon-based energy source, and its thermochemical conversion into hydrogen can potentially achieve near-zero lifecycle carbon emissions [3]. Among the available routes, biomass gasification coupled with catalytic reforming has attracted considerable attention because of its feedstock flexibility and relative process maturity. Nevertheless, syngas derived from conventional biomass gasification generally suffers from low hydrogen content (typically 20-40 vol% on a dry basis), high tar concentrations (10-50 g Nm^{-3}), and the presence of various impurities. Catalytic reforming can significantly enhance hydrogen yield and improve syngas quality by promoting tar cracking, the water-gas shift reaction, and methane reforming [4].

Catalysts are the decisive factor governing the efficiency of the gasification-reforming process. An ideal catalyst should exhibit high catalytic activity, strong resistance to coke deposition and sintering, robust mechanical strength, and favorable economic viability. At present, a variety of catalyst systems have been explored for biomass reforming-based hydrogen production, including naturally occurring minerals such as dolomite and olivine, transition metals such as Ni, Fe, and Co, noble metals such as Rh, Ru, Pt, and Pd, alkali and alkaline earth metals such as Na, K, Ca, and Mg, as well as emerging bifunctional materials that integrate CO_2 adsorption and/or oxygen carrier functions [5-7]

This review focuses on three core aspects, namely catalyst systems, key operating parameters, and catalyst deactivation mechanisms, with the aim of systematically summarizing recent progress in biomass gasification-catalytic reforming for hydrogen production.

2. Fundamentals of Biomass Gasification and Catalytic Reforming

2.1 Major Gasification and Reforming Reactions

Biomass gasification is a thermochemical conversion process in which solid biomass is transformed into combustible gases under elevated temperatures, typically in the range of 600-1000 °C, and a controlled oxidizing atmosphere, such as air, O_2, steam, CO_2, or their mixtures [8]. The principal reactions are summarized in Table1. Overall, the process can be simplified as follows:

Biomass → char + tar + gases (H_2, CO, CO_2, CH_4) + H_2O

Table 1: Key reactions in biomass gasification and reforming

Reaction name	Reaction equation	Reaction enthalpy (kJ mol^-1)	No.
Steam gasification	$C+H_2O \rightarrow CO+H_2$	-118.9	R1
Boudouard reaction	$C+CO_2 \rightarrow 2CO$	-173.78	R2
Methanation	$C+2H_2 \rightarrow CH_4$	+74.84	R3
Water-gas shift reaction	$CO+H_2O \rightarrow CO_2+H_2$	+40.9	R4
Steam reforming of methane	$CH_4+H_2O \rightarrow CO+3H_2$	-206.3	R5
Dry reforming of methane	$CH_4+CO_2 \rightarrow 2CO+2H_2$	-247.3	R6
Reforming of hydrocarbons	$C_nH_m+H_2O \rightarrow nCO+(n+\frac{m}{2})H_2$	--	R7

Table 2: Representative Ni-Based Catalysts and Their Performance in Biomass Gasification and Reforming

Catalyst Formulation	Feedstock	Reaction Conditions	H_2 Yield (vol%) or Productivity	Key Features	Ref.
10% Ni/Al_2O_3	Pine sawdust	800 °C, S/C = 1.5	~52 vol% H_2 in syngas	High initial activity	[15]
$Ni/CeO_2–Al_2O_3$	Cellulose	750 °C, steam	~60 vol% H_2	Enhanced oxygen mobility	[16]
Ni–Fe/olivine	Rice husk	850 °C, S/C = 1.0	55-62 vol% H_2	Bimetallic synergy	[17]
Ni–Co/dolomite	Wood chips	800 °C, air/steam	~58 vol% H_2	Improved coke resistance	[18]
$Ni/MgO–Al_2O_3$	Corn stalk	750 °C, S/C = 1.2	64 vol% H_2	Spinel suppression	[19]

2.2 The Core Role of Catalysts

Catalysts play three central roles in biomass gasification: first, they substantially reduce the activation energy required for cleavage of the C-C and C-O bonds in tar macromolecules, thereby accelerating tar cracking reactions; second, they promote the water-gas shift reaction and facilitate the conversion of CO into H_2, leading to higher hydrogen yields; and third, they suppress excessive tar formation and carbon deposition, thereby alleviating downstream equipment blockage. Compared with non-catalytic gasification, the use of an appropriate catalyst can increase H2 yield by 30% to 100% [9]. Yao et al [10] systematically investigated the catalytic performance of biochar-supported nickel catalysts and found that nickel catalysts supported on different biochars all exhibited favorable catalytic activity in gasification-based hydrogen production, providing important guidance for the design of low-cost catalysts.

3. Catalyst Systems

3.1 Nickel-Based Catalysts

Nickel-based catalysts are the most extensively investigated and technologically mature non-noble metal catalyst systems. They exhibit high activity for C-C and C-H bond cleavage and are far less expensive than noble metals [9]. Their catalytic performance is mainly determined by the choice of support, metal loading, promoter type, and preparation method. Commonly used supports include γ-Al_2O_3, SiO_2, MgO, CeO_2, ZrO_2, and zeolites. Although Al_2O_3 offers a high specific surface area of 150–300 m_2/g, it is prone to forming $NiAl_2O_4$ spinel at elevated temperatures, thereby reducing catalytic activity [11]. In contrast, basic or reducible supports such as MgO and CeO_2 can effectively suppress sintering through strong metal-support interactions with nickel.

The incorporation of alkali and alkaline earth metals such as K, Mg, and Ca can modify the surface acidity and basicity of the catalyst. Rare-earth oxides such as CeO_2 and La_2O_3 promote carbon gasification through their oxygen storage capacity, while transition metals such as Co, Fe, and Cu are often introduced to form bimetallic systems and generate synergistic effects [12]. Wu et al. [13] systematically investigated the deactivation mechanisms of Ni-based catalysts in biomass gasification tar steam reforming and found that carbon deposition and thermal sintering were the primary causes of catalyst deactivation. The Ni/Ca_3AlO catalyst developed by Huo et al. [14] achieved a hydrogen yield of 30.08 mmol/g biomass and a hydrogen concentration of 60-61 vol% in the first reaction cycle. After 10 regeneration cycles, only slight carbon deposition and minor sintering were observed, demonstrating excellent cycling stability.

3.2 Iron-Based Catalysts

Iron-based catalysts are highly cost-effective and environmentally benign, and they show outstanding performance in chemical-looping gasification systems and the water-gas shift reaction. The valence state of iron has a significant influence on catalytic activity: metallic Fe exhibits high activity for tar reforming but is prone to oxidation and deactivation under steam-rich conditions; Fe_3O_4 shows moderate activity for the water-gas shift reaction; and Fe_2O_3 has relatively good stability but lower activity [20].

Yang's group [21] systematically investigated the effect of CeO_2 addition on the hydrogen production performance of Fe-based catalysts in cellulose steam gasification. They examined the catalytic behavior at different Ce/Fe molar ratios (0:1, 3:7, 5:5, 7:3, and 1:0). The results showed that the catalyst with Ce/Fe = 5:5 exhibited the best performance, with a significant increase in hydrogen yield, which was attributed to the enhanced carbon gasification efficiency arising from the oxygen storage capacity of CeO_2. The Xiao Rui research group at Southeast University has also carried out systematic work on the development of iron-based oxygen carriers for chemical-looping hydrogen production. In chemical-looping gasification, iron-based oxygen carriers partially oxidize biomass in the fuel reactor to produce nitrogen-free syngas, and are then regenerated in the air reactor. The resulting syngas can be further reacted with steam to release high-purity hydrogen.

3.3 Noble Metal Catalysts

Noble metals such as Rh, Ru, and Pt exhibit extremely high catalytic activity for tar cracking and the water-gas shift reaction, and they generally outperform nickel-based catalysts, especially at relatively low temperatures of 450–650 °C [22]. However, their high cost severely limits large-scale application. Recent research has focused on single-atom catalysts (SACs) and bimetallic catalysts with low noble-metal loading to maximize atomic utilization. Pt-based alloy catalysts can significantly improve selectivity and stability by tailoring the surface atomic arrangement.

3.4 Alkali and Alkaline Earth Metal Catalysts

Alkali metals such as K and Na, as well as alkaline earth metals such as Mg and Ca, are naturally present in biomass ash. K_2CO_3 and Na_2CO_3 promote the cleavage of C-O bonds by disrupting aromatic ring structures, thereby accelerating tar reforming and char gasification [23]. CaO has a dual function: it catalyzes the water-gas shift reaction and captures CO_2 in situ. Li et al. [24] systematically studied the key parameters affecting CaO-based sorption-enhanced steam gasification using a thermodynamic equilibrium model. The results showed that increasing the temperature from 823 K to 923 K and increasing the steam-to-biomass mass ratio from 0.5 to 1.5 improved both the H_2 concentration and gas yield, whereas increasing the CaO-to-carbon molar ratio from 1 to 2 mainly enhanced the hydrogen concentration. However, alkali and alkaline earth metal catalysts are prone to volatilization or loss at high temperatures above 750 °C, and regeneration of CaO-based sorbents requires high-temperature calcination above 900 °C, resulting in relatively high energy consumption.

3.5 Bifunctional Catalysts and Biochar-Based Catalysts

Bifunctional catalysts integrate reforming activity with in situ CO2 capture in a single material, representing a frontier direction for high-purity hydrogen production and negative-carbon-emission processes. Yang's group [25] employed electrically driven flash pyrolysis of biomass, using joule heating to rapidly raise the biomass temperature to above 2000 °C within seconds, thereby completing pyrolysis and in situ cracking of volatiles. This technique achieved superior performance compared with conventional methods in terms of hydrogen efficiency (51.6%), syngas quality (H_2/CO ≈ 1:1), and carbon material properties, and its environmental and economic benefits were validated through life-cycle assessment and techno-economic analysis.

Biochar-based catalysts utilize the intrinsic pore structure, surface functional groups, and mineral species of biochar as active sites or supports. Park et al. [26] found that Ni-impregnated biochar exhibited catalytic activity comparable to that of Ni/γ-Al_2O_3, while the spent catalyst could be recovered by combustion for heat generation, eliminating secondary solid waste. In addition, Ni-supported hydrochar catalysts have shown good tar removal efficiency in low-temperature tar reforming. Zhang et al. [27] developed a carbon-supported NiFe–$NiFe_2O_4$ catalyst for low-temperature catalytic cracking of biomass tar. This study provides a feasible technical route for the application of biochar-based catalysts under low-temperature conditions.

4. Effects of Key Operating Parameters

Gasification temperature, steam-to-biomass ratio, catalyst dosage, equivalence ratio, and other operating parameters directly affect reaction equilibrium, reaction kinetics, and hydrogen yield. Appropriate parameter regulation is therefore essential for efficient hydrogen production.

4.1 Gasification Temperature

Temperature is a core parameter in biomass gasification and reforming, and it plays a decisive role in both reaction kinetics and equilibrium distribution. Increasing the temperature favors endothermic steam reforming and tar cracking reactions, thereby improving hydrogen yield and reducing tar content. Ji et al. [28] systematically investigated the effects of gasification temperature, steam-to-biomass mass ratio (S/B), and equivalence ratio on product gas composition and hydrogen production. The results showed that at a gasification temperature of 800 °C, the hydrogen volume fraction increased with increasing S/B or decreasing equivalence ratio, and the hydrogen yield reached a maximum at either S/B = 1.5 or an equivalence ratio of 0.22. When the temperature was raised to 950 °C, with S/B = 1.5 and an equivalence ratio of 0.22, both the hydrogen volume fraction and yield reached their maximum values of 35.47% and 78.22 g/kg, respectively.

However, excessively high temperatures also bring adverse effects. Lysne et al. [29] studied the steam reforming performance of simulated biomass gasification tar impurities using a Ni-Co/Mg(Al)O catalyst. They found that although higher temperatures promoted tar conversion, they also accelerated carbon deposition and catalyst sintering. Their experiments were conducted systematically at different temperatures (650-800 °C), steam-to-carbon ratios (2-5), tar loadings (10-30 g Nm^{-3}), and tar compositions. The catalyst deactivated due to carbon deposition after complete tar removal.

4.2 Steam-to-Biomass Ratio and Gasifying Agent Type

Steam plays a dual role in gasification systems: it participates as a reactant in char gasification and tar reforming reactions, and it enhances hydrogen selectivity through the water-gas shift reaction. However, excessive steam increases energy consumption because of its high latent heat of vaporization, and it may also cause bed agglomeration in fluidized-bed systems.

Ji et al. [28] showed that at a gasification temperature of 800 °C, the hydrogen volume fraction increased with increasing S/B, and the H2 yield reached a maximum at S/B = 1.5. In the temperature range of 750–950 °C, increasing both the gasification temperature and S/B led to a continuous increase in H2 concentration, along with corresponding increases in hydrogen yield and steam conversion. Through an orthogonal experimental design, they analyzed the effects of individual factors and their interactions on hydrogen concentration and found that the equivalence ratio had the most significant influence, followed by gasification

temperature and steam-to-biomass mass ratio. The optimal operating conditions were a gasification temperature of 800 °C, a steam-to-biomass mass ratio of 2.5, and an equivalence ratio of 0.22, under which the hydrogen concentration reached 38.27%.

4.3 Catalyst Dosage and Ca/C Molar Ratio

For calcium-based sorption-enhanced systems, the amount of catalyst or sorbent directly affects hydrogen yield and CO_2 capture efficiency. Studies have shown that the suitable range of the Ca/C molar ratio is 1.0-2.0. As the Ca/C molar ratio increases to 2.0, the H_2 volume fraction can rise to 60.5%-67.8%, while the CO_2 volume fraction can decrease to 0.2%-8.5%; the CH4 content remains essentially within 10%-17%.

Zhu et al. [30] investigated the co-gasification and steam reforming of biomass and plastic mixed wastes and found that an appropriate amount of alkali metal addition could significantly promote tar cracking and improve hydrogen yield. In addition, biomass internal and external metal synergistic catalytic gasification for hydrogen production has achieved efficient tar reforming and enhanced hydrogen selectivity through the cooperative distribution of catalysts inside and outside the biomass structure.

5. Catalyst Deactivation Mechanisms and Suppression Strategies

Catalyst deactivation is the key bottleneck limiting the industrialization of catalytic biomass gasification reforming. The high-temperature conditions above 700 °C, the complex tar composition, and the presence of sulfur- and chlorine-containing impurities in biomass gasification expose catalysts to the combined effects of multiple deactivation pathways.

5.1 Carbon Deposition Formation and Classification

Carbon deposition is the most common deactivation mode for Ni-based catalysts. It is mainly formed through hydrocarbon decomposition, which generates filamentous or graphitic carbon, and the Boudouard reaction, $2CO \rightarrow C + CO_2$, which produces amorphous carbon. Lysne et al [29]. systematically characterized carbon deposition on Ni-Co/Mg(Al)O catalysts and proposed a classification scheme, including surface adsorbed carbon (Type A), initial carbon filaments (Type B1), fused carbon filament clusters (Type B2), and strongly deactivating encapsulating carbon (Type B3). High-molecular-weight tar is more likely to form encapsulating carbon that is difficult to remove. Carbon deposition not only covers active sites but also blocks pores and increases internal diffusion resistance.

5.2 Metal Sintering and Impurity Poisoning

At high temperatures of 700–950 °C, metal nanoparticles migrate and coalesce, leading to sintering and irreversible deactivation. Jin et al. [31] pointed out that sintering and redispersion during regeneration can cause permanent activity loss. In addition, sulfur and chlorine impurities in biomass can poison catalysts: sulfur forms stable sulfides, while chlorine generates metal chlorides, resulting in the loss of active components.

5.3 Main Strategies for Suppressing Deactivation

1) Bimetallic synergistic design. Introducing a second metal such as Co, Fe, or Cu to form bimetallic catalysts can alter the electronic structure of Ni, weaken the tendency for C-C bond dissociation to generate active carbon species, and reduce surface acidity/basicity to suppress deep dehydrogenation reactions. Lysne et al. [29] showed that the basic sites of the Mg(Al)O support can assist the gasification of coke and its precursors, thereby suppressing carbon accumulation.

2) Promoter introduction and surface modification. Oxide promoters such as CeO_2 and La_2O_3 can promote carbon gasification through their oxygen storage and release capacity, producing an in situ regeneration effect. Yang's group [21] found that adding CeO_2 to Fe-based catalysts significantly improved carbon-resistance performance because the oxygen storage capability of CeO_2 enhanced carbon gasification efficiency.

3) Optimization of regeneration methods. Proper selection of the regeneration atmosphere and conditions can effectively restore catalyst activity. Azancot et al. [32] found that Ni–K catalysts could be fully regenerated in air and CO_2 atmospheres, whereas Ni catalysts without K were difficult to reactivate. Gai et al. [33] proposed a simple N_2 regeneration treatment (800 °C, 30 mL/min, 30 min), which restored the tar reforming efficiency of a deactivated Ni-loaded hydrochar catalyst to 71%, with H_2 selectivity reaching 38% and an H_2 yield of 758 mL/g, significantly outperforming conventional CO_2, O_2, or air regeneration methods. This N_2 treatment suppressed excessive oxidation of the coke/hydrochar support while inducing redispersion of Ni species into smaller nanoparticles, thereby achieving both carbon removal and inhibition of metal sintering.

6. Conclusions

This review systematically summarizes recent progress in hydrogen production via catalytic biomass gasification reforming, focusing on three core topics: catalyst systems, key operating parameters, and catalyst deactivation mechanisms. Through comparative analysis of nickel-based, iron-based, noble metal, alkali/alkaline earth metal, bifunctional, and biochar-based catalysts, the applicable scenarios and improvement directions of different systems were clarified. Ni-based catalysts, due to their combined advantages in activity and cost, show the greatest short-term commercialization potential. Iron-based oxygen carriers exhibit unique cost and cyclic performance advantages in chemical-looping hydrogen production. Alkali and alkaline earth metal catalysts are suitable for sorption-enhanced processes, whereas biochar-based catalysts provide a low-cost and sustainable option for small-scale distributed applications.

At the process level, gasification temperature, steam-to-biomass ratio, and catalyst dosage exhibit complex interactive coupling effects, and global optimization is more conducive to efficient hydrogen production than stepwise

adjustment. The dominant deactivation mechanisms of catalysts remain carbon deposition and sintering. Carbon deposition evolves through multiple stages, from surface adsorbed carbon to carbon filaments and further to graphitized encapsulating carbon, and high-molecular-weight tar significantly accelerates this process. In response to these deactivation pathways, this review summarizes suppression strategies such as bimetallic synergistic design, core-shell confinement, promoter introduction, and optimized regeneration. Rational combination of these strategies is expected to significantly extend catalyst lifetime in industrial applications. Overall, this review presents the latest progress in biomass gasification reforming for hydrogen production and highlights future research directions, including catalyst design, process intensification and carbon-negative technology integration, as well as lifecycle assessment of sustainable catalysts.

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