



Preparation, modification and development of Ni-based catalysts for catalytic reforming of tar produced from biomass gasification

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ABSTRACT

Biomass gasification is one of the most promising technologies to convert renewable biomass feedstock to useful energy and chemicals for decarbonizing the current industrial activities. However, complex tar compounds are formed in the produced syngas. The presence of tar in syngas is undesirable due to a series of problems caused, such as the decrease of overall efficiency and the clogging and contamination of downstream equipment. Until now, catalytic steam reforming has been widely studied for the efficient removal of tar. Previous review articles have focused on the catalytic reforming of tar by categorizing various catalysts as basic catalysts, nickel-based catalysts, non-nickel based catalysts, alkali metal catalysts, zeolite catalysts, and carbon-supported catalysts, etc. Ni-based steam-reforming catalysts have attracted much attention due to their high activity for tar reduction, low-cost and easy regeneration. However, the deactivation caused by the coke deposition and metal sintering remains the greatest challenge for the deployment of the technology. Therefore, modified Ni-based catalysts are now most frequently used for catalytic reforming of tar. At present, few review articles reported the modification of Ni-based catalysts for catalytic reforming of tar compounds. Based on this, the preparation, modification and development methods of Ni-based catalysts for catalytic reforming of tar are reviewed in this paper, aiming at promoting the catalytic performance of conventional Ni-based catalysts, in terms of high catalytic activity, long-term stability and better selectivity towards low molecular weight compounds.

1. Introduction

1.1. Biomass energy

Renewable energy sources (RES), including biomass, hydropower, geothermal, solar, wind and marine energies, provide 14% of the total world energy demand [1]. Among the RES, biomass energy derived from agricultural, forest and municipal solid wastes is considered as one of the most promising alternatives [2], due to its abundant resources and neutral property in CO₂ circulation. According to the global renewable energy scenario by 2040 (Table 1), the share of biomass is

much more than other RES, contributing more than 50% of the total RES [1]. In addition, bioenergy has various social and environmental benefits, such as reducing the emission of greenhouse gases (GHGs), NO_x and SO_x, lowering the risk of soil and water acidification, increasing the value of agricultural output, and decreasing the dependence on fossil energy [3]. The increasing consumption and high price of fossil energy (coal, oil and natural gas), together with the rising threat of global warming, make the utilization of biomass more and more attractive [4]. Biomass energy has been the fourth largest energy source since 1990, and occupies 13% of the world's total primary energy consumption (55 EJ per year) (Fig. 1). Specifically, a much larger

Abbreviations: RES, renewable energy sources; GHGs, greenhouse gases; RIGES, renewable intensive global energy scenario; DME, dimethylether; PAHs, polycyclic aromatic hydrocarbons; HMA, hexamethylenetetramine; REOs, rare earth oxides; WGS, water-gas shift reaction; NTP, non-thermal plasma; OSC, oxygen storage capacity; CS, calcined scallop shell; OCs, oxygen carriers; NFA, nickel-iron alloy catalysts; 1-MN, 1-methylnaphthalene; NM, noble metals; AIMD, ab initio molecular dynamic; SMNi, Ni-incorporated mesoporous smectite-like material; AAEM, alkali and alkaline earth metals; HTLcs, hydrotalcite-like compounds; SEM, scanning electron microscope; XRD, X-ray powder diffraction; TPR, temperature-programmed reduction; TPD, temperature programmed desorption; TGA, thermogravimetry; XPS, X-ray photoelectron spectroscopy

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Table 1
Global renewable energy scenario by 2040 [1].

	2001	2010	2020	2030	2040
Total consumption (million tons oil equivalent)	10,038	10,549	11,425	12,352	13,310
Biomass	1080	1313	1791	2483	3271
Large hydro	22.7	266	309	341	358
Geothermal	43.2	86	186	333	493
Small hydro	9.5	19	49	106	189
Wind	4.7	44	266	542	688
Solar thermal	4.1	15	66	244	480
Photovoltaic	0.1	2	24	221	784
Solar thermal electricity	0.1	0.4	3	16	68
Marine (tidal/wave/ocean)	0.05	0.1	0.4	3	20
Total RES	1365.5	1745.5	2964.4	4289	6351
Renewable energy source contribution (%)	13.6	16.6	23.6	34.7	47.7

proportion of 33% is obtained for biomass in developing countries, containing 75% of world's population [5], because agriculture is a major industry in developing countries [6]. In addition, it is estimated in the renewable intensive global energy scenario (RIGES) in 1992 that, by 2050, biomass could provide half of the world's total primary energy consumption (400 EJ per year), and almost 60% of electricity demand in the world could be supplied by renewables among which biomass is an important feedstock [7].

Biomass is mainly composed of cellulose, hemicellulose, lignin and minerals [8]. Biomass can be utilized to produce heat, electricity and various chemical feedstock by various methods including thermal-chemical processes and bio-chemical processes. The thermal-chemical processing of biomass includes direct combustion, pyrolysis and gasification [9]. Among them, biomass gasification is one of the most economical and efficient methods to convert biomass into energy species [10,11].

1.2. Biomass gasification

Gasification of biomass is a thermal-chemical process where carbonaceous materials can be converted to combustible gas fuel in the presence of limited amount of oxidizing agents (e.g. O_2 , air, steam, CO_2) at relative high temperatures ($> 700^\circ C$). [10,11] On one hand, the processing of biomass wastes is beneficial to the reduction of landfill disposal and global warming [12]. On the other hand, the obtained syngas from biomass gasification can be used as secondary raw feedstock to produce methanol, dimethylether (DME), ethanol, Fischer-Tropsch fuels, as well as generating heat and electricity [13]. Meanwhile, some undesirable by-products, such as tar and solid particulates, are also generated [14]. The main processes and reactions of biomass gasification are shown in Fig. 2. It is indicated that biomass gasification can be generally classified into four steps: drying, pyrolysis, oxidation, and gasification [15]. The exceeded moisture should be evaporated firstly upon heating the wet solid biomass to around $200^\circ C$ to generate dry fuel. Pyrolysis mainly occurs in the temperature range of $150\text{--}900^\circ C$, where the cellulose, hemicellulose and lignin are decomposed into gases (such as CO , CO_2 and CH_4), condensable liquids and carbon-rich char. The condensable liquid of "Tar" contains complex and various organic and inorganic compounds. The oxidation is a significant process occurring at more than $700^\circ C$, where the pyrolysis products (such as CO , CH_4 and C) are oxidized to CO , CO_2 , H_2O , etc in the presence of oxidants (e.g. air and H_2O). The main reactions during oxidation process are the endothermic oxidation reactions of pyrolysis products, as shown in Fig. 2. In general, within the gasification process, a series of reactions, such as cracking, reforming, condensation, polymerization, oxidation and gasification reactions, take place in the presence of air, steam and CO_2 [16,17]. The final products of the entire biomass gasification process contain synthetic gas (such as H_2 , CO , CO_2

and CH_4), condensable tar, and solid char [18].

Evans and Milne [19] classified the main processes of biomass gasification as the primary process (primary oxygenates), secondary process (hydrocarbon) and tertiary process (large aromatic). The conversion pathway of biomass gasification in the solid, liquid and vapor phases are presented in Fig. 3. During the primary process, solid biomass is converted to primary oxygenated vapors, primary liquids and gaseous H_2O , CO and CO_2 in the temperature range of $400\text{--}700^\circ C$. The primary oxygenated vapors are of lower molecular weight, including the monomers and monomer fragments of the biopolymers, such as levoglucosan, furfural, hydroxyacetaldehyde and methoxy phenols [20]. Only slight cracking reactions occur to generate lighter aromatics and oxygenates, and no chemical interactions are observed between the organic components of biomass during primary process.

During the secondary process, light olefins, aromatics, and gaseous H_2O , H_2 , CO and CO_2 are generated from the cracking of methoxy phenols at $700\text{--}850^\circ C$. In addition, the primary liquids form condensable oil that contains phenol and aromatics. For the tertiary process, the further increase of temperature to above $850\text{--}1000^\circ C$ leads to the continuous formation of polycyclic aromatic hydrocarbons (PAHs), as well as CO , H_2 , CO_2 and H_2O through the high-temperature conversion process such as gasification and combustion. The tar compounds generally contain benzene, toluene, indene, naphthalene, anthracene, phenanthrene, and pyrene that are derived from the polymerization of the hydrocarbons with low molecular weight. In addition, the condensation of these tar compounds results in the formation of a liquid mixture at low temperatures.

In addition to the gaseous and liquid products, different kinds of solid products are also generated through different processes and defined on the basis of their origins. For example, charcoal is produced in the primary process and maintains the property of original lignocellulose. Both coke and soot are formed in the secondary and tertiary processes, where coke is derived from the thermolysis of the depositional liquid and organic vapors, and soot is produced from the homogeneous nucleation of the intermediates decomposed from the hydrocarbons in the vapor phase at high temperatures.

1.3. Tar issues

Among the products derived from biomass gasification, the presence of tar in syngas is one of the most critical barriers limiting the commercial application of biomass gasification. Tar is a complex mixture of condensable organic compounds including single to multiple ring aromatic compounds, along with oxygenates and heavy PAHs [21,22]. The content of tar varies from 1.0 to 100 g/m^3 depending on gasifier type, feed type and operation parameters, etc. [23]. The formation of tar will cause several problems, such as the loss of chemical energy in syngas, soot formation, and the decrease of overall energy efficiency [24,25]. In addition, tar can condensate and polymerize to form heavier structures, leading to the contamination and plugging of pipe, engines, turbine, filter and heater exchanges, and the poison of catalysts during biomass gasification and biogas utilization [26–28]. Therefore, tar content generally should be controlled below 1.0 g/m^3 for the commercial application of fuel gas [29]. Based on this, the purification of fuel gas and the destruction of tar are necessary for the application of gasification technologies [30].

1.4. The objectives of this review

Based on the above analysis, the efficient and economic removal of undesirable tar in biomass gasification are reviewed in this work, aiming at giving guidance for future studies towards the commercial application of biomass gasification. Until now, catalytic steam reforming of tar has been widely reviewed in similar papers. For example, previous articles focused on reviewing different technologies for tar removal, such as physical methods, thermal removal, catalytic

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