



Research paper

Elucidation of syngas composition from catalytic steam gasification of lignin, cellulose, actual and simulated biomasses



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ABSTRACT

This work presents research into the gasification of cellulose, lignin, simulated and two actual biomasses which are rich in cellulose and lignin contents, in order to evaluate and compare their gas products and also understand their gasification processes in presence of CaO by varying the CaO to biomass feed ratio from 0 to 1.5 at 700 °C. Biomass components at the optimum CaO loading were gasified at different temperatures ranging from 600 to 800 °C, and then actual and simulated biomasses were gasified at optimum observed temperature. It was observed that increasing the CaO/cellulose ratio from 0 to 1.5 enhanced the H₂ improvement. In contrast, the hydrogen yield in lignin gasification slightly reduced when CaO/lignin mass ratio was more than 1; this could be due to (i) lower content of sodium (Na) in the gasified sample at CaO/lignin ratio 1.5 and (ii) interaction of Ca(OH)₂ with H₂ molecules with increasing the Ca atoms. Maximum H₂ concentration was achieved with their corresponding CaO loading ratio at 650 °C in both the components. The selected biomass components not only have an effect on product gases but also have a significant influence on the catalyst activity. The gas evolution trend from actual and simulated biomasses results was similar to those selected biomass components. Thus, the higher cellulose content of the biomass, the higher would be the CO yield. Correspondingly, the higher the lignin percentage of the biomass, the higher could be the CH₄, CO₂, and H₂ yields.

1. Introduction

As a result of environmental and economic incentives such as rising energy prices and the depletion of fossil fuels, countries are changing their energy profiles towards renewable and sustainable resources [1]. Many technologies based on alternative energy sources have emerged. One of the technologies that is gaining in importance is the gasification of biomass [2]. Gasification is a key technology for the clean and efficient usage of coal, biomass and other carbonaceous materials [3]. Gasification is a thermal process in which a material, such as biomass, is exposed to a low-oxygen environment and undergoes a sequence of two endothermic transformations. The biomass decomposes during the first conversion at about 600 °C into volatiles and char. The resulting gas contains hydrocarbon gases, methane, hydrogen, carbon monoxide, carbon dioxide, tar, water vapor and nitrogen [4]. The gasification agent is selected according to the desired chemical composition of the synthesis gas and the efficiency [5]. The production of H₂ from biomass gasification can be considered as an advanced exploitation of this traditional energy resource, with the potential to play an important role in sustaining a futuristic H₂ energy economy [6,7]. In fact, biomass contains varying amounts of cellulose, hemicellulose, lignin and few other

extractives [8]. The amounts of cellulose, hemicellulose, and lignin vary substantially with the type of biomass materials [9] which have rough formulae as CH_{1.67}O_{0.83}, CH_{1.64}O_{0.78}, and C₁₀H₁₁O_{3.5}, respectively [10]. Chemical differences in the structural components of the biomass (cellulose, hemicellulose, and lignin) have a direct impact on its chemical reactivity [11] which affect the characteristics of the gasification process [12]. Typically, cellulose, hemicellulose, and lignin constitute about 85–90% of lignocellulosic biomass; organic extractives and inorganic minerals constitute the rest [13]. Cellulose and lignin are widely recognized as the major constituents of biomass, which are some of the important parameters for the evaluation of pyrolysis and gasification properties [14]. Lignin fraction normally consists of 20–40 wt% dry of biomass. Lignin is a complex racemic polymer and is composed of p-hydroxyphenyl, guaiacyl and syringyl units; an example of a lignin structure is shown in Fig. 1A. Since only the lignin fraction of the biomass is aromatic in nature, lignin represents a potential precursor for PAH formation. Three hydroxycinnamyl alcohols considered precursors of lignin, which only differ in their degree of methoxylation [15]. In contrast, Cellulose is a linear homopolysaccharide of cellobiose monomers, composed of two β-glucopyranose units as presented in Fig. 1B. All the covalent linkages between the β-glucopyranose motifs

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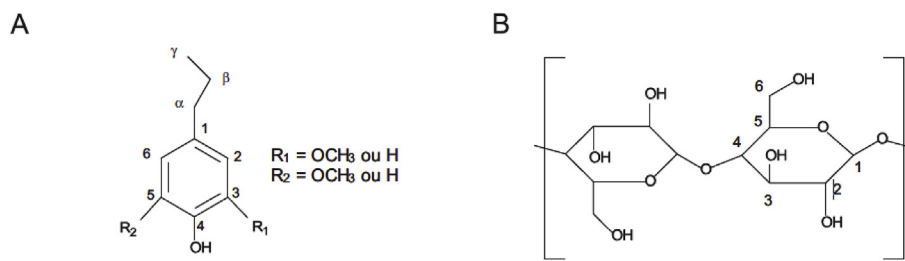


Fig. 1. Chemical structures of biomass constituents (A) Building units of lignin; (B) building units of cellulose [16].

of the framework are β -1,4-glycosidic bonds [16]. Generally speaking, the lignin is harder to decompose than the cellulose due to the part of lignin in the biomass consists of benzene rings [17].

Several researchers examined the effect of biomass component on the pyrolysis process and studied the decomposition behavior of the components [8,18–21]. Burhenne et al. [22] identified characteristic differences in the pyrolysis behavior of three widespread biomass sources from biomass sources, which differ in cellulose, hemicellulose and lignin fractions. It is shown that the lignin content of each biomass feed is the major factor in the control of pyrolysis in industrial processes. Higher lignin content leads to a slower decomposition, a lower product gas yield and a higher temperature. Couhert et al. [23] investigated for the prediction of pyrolysis gas yields from any biomass on the basis of its composition in cellulose, hemicellulose, and lignin. They established a link between the composition of a biomass and its pyrolysis gas yields and composition. An attempt was made to predict gas yields of any biomass according to its composition.

In addition to the studies on the pyrolysis of biomass components described above, Some researchers have examined the steam catalytic gasification of different biomass components for the production of hydrogen. Wu et al. [24] investigated the hydrogen and syngas production from the pyrolysis/gasification of biomass components (cellulose, xylan, and lignin) in the presence of Ni-based catalysts by using a two-stage fixed-bed reaction system. Biomass samples were pyrolysed at the first stage and the derived products were gasified at the second stage. Their work shows that the components of biomass have a significant influence on the catalytic gasification process related to hydrogen and syngas production. In another their study [25], they found that when Ni-Zn-Al (1:1) catalyst was used in the gasification process, gas yield was increased from 62.4 to 68.2 wt% for cellulose, and from 25.2 to 50.0 wt% for the pyrolysis/gasification of lignin. Inaba et al. [26] investigated the hydrogen production by cellulose gasification over Ni catalysts. The catalytic activity and the species of products were affected by the type of supports.

Some researchers in this field have focused on using different catalysts (ZrO_2 , NaOH, Pt, RuO_2 , Ru, Ni) [24,25,27–32]. However, the investigation of steam gasification of biomass components, such as cellulose and lignin, in presence of pure CaO has rarely been investigated.

CaO is low cost and abundant because it can be derived from a range of naturally occurring precursors including limestone, dolomite and calcium hydroxide [33,34]. During the gasification process, CaO is added into the reactor, to react with CO_2 to produce solid $CaCO_3$ in

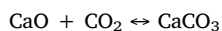


Table 1
Ultimate and proximate analysis of the selected biomass components.

Biomass	Ultimate analysis (wt.%)				Proximate analysis (wt.%)			
	Carbon	Hydrogen	Oxygen	Nitrogen	Fixed Carbon	Volatile Matter	Moisture	Ash
Cellulose	50.6	5.35	43.95	0.1	0.45	95.36	3.9	0.29
Lignin	64.19	6.22	29.58	0.01	34.34	47.88	4.93	12.85

The reaction above is called carbonation, which is readily reversed by the application of heat [35] and plays two important roles: (1) absorbing the released CO_2 [36–39] to enhance the water gas and water gas shift reactions towards hydrogen production due to their high stability in the carbonation and calcination (regeneration) [6,33,37,40]; (2) providing necessary energy for the endothermic gasification through the carbonation reaction releasing heat [41,42]. Another potential advantage of gasification enhanced by a CO_2 sorbent is the possibility to reduce tars in the syngas by catalysis of the CaO additives [39,43–46].

Wei et al. [47] investigated the characteristics of steam gasification of biomass for hydrogen production at ambient pressure in a laboratory scale external circulating concurrent moving bed (ECCMB) system with CaO as a CO_2 absorbent. Their results showed that the addition of a CaO based CO_2 absorbent is a good solution to increase hydrogen content in dry gas from the ECCMB process. Gas yields and carbon conversion efficiencies have been studied for the steam gasification of white fir [48], sawdust [49] and pine bark [50] by using CaO catalyst. Hanaoka et al. [42] examined the operation conditions for H_2 production from Japanese oak by steam gasification using CaO as a CO_2 sorbent. Although, these researchers have used typical biomass materials to study the gasification process in presence of CaO. But an understanding of the relationship between the biomass components (Cellulose and Lignin) and CaO performance is still lacking. Therefore, CaO was selected to understand and compare the performance of both components in same operating conditions, and to estimate the effect of CaO on components, while concentrating specifically on the H_2 gas production from gasification process.

2. Experimental setup

2.1. Feeding materials and catalyst

Lignin and cellulose were used in this research as model components and both the feeding materials were purchased from Shanghai Macklin Biochemical and Tokyo Chemical Industry Co., Ltd respectively. Moreover, Corn Stover and walnut shell were employed as real biomass. The ultimate and proximate analyses of components are shown in Table 1. Calcium oxide (CaO) was calcined at $950^\circ C$ for 3 h before it was used. The purpose of using pure CaO as in bed-catalyst was to prevent the interference of various impurities on the catalytic activity of CaO.

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