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Analysis of the catalytic steam gasification mechanism of biomass

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ABSTRACT

The gasification of cellulose was here used to prove that the inner reaction of oxygen with carbon contained in the biomass was unavoidable during the gasification of biomass. It was also proven that the dehydrogenation reaction and conversion of oxygen in biomass to CO always caused the formation of residual carbon. Although, almost all of oxygen contained in the biomass could be converted into CO by the catalytic effect of ions, which in turn could be converted to CO₂, but the CO₂ was always methanized. This indicated that the hydrogen and syngas yield contained in the product gas of biomass could not practically achieve to the theoretical maximum hydrogen and syngas yield from biomass. The present study showed biomass gasification connected in series with coal gasification to be an excellent approach to large-scale conversion of biomass to hydrogen and syngas.

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Introduction

Hydrogen and syngas are clean energies and important raw materials in industry. Hydrogen is likely to be an important energy carrier in the future. Carbonaceous fossil fuels are currently the most common source of hydrogen and syngas; however, these fuels do not meet the requirements for sustainable development. Biomass is a source of abundant, environmentally friendly, and renewable energy, and it may be an ideal alternative to fossil fuels for hydrogen and syngas [1–6].

Many typical techniques, such as biological conversion, pyrolysis, catalytic pyrolysis, supercritical water gasification, (catalytic) air and oxygen/steam gasification, and catalytic steam gasification from biomass have been developed to

produce hydrogen-rich gas, syngas, and hydrogen [4,7–13]. Catalytic steam gasification of biomass is the most promising of these techniques [14,15].

This technique has several advantages: It increases the reaction rate of steam with biomass and participates in the secondary reactions. It can adjust tar levels and the resulting gas composition, producing a gaseous fuel with a relatively high H₂ content, eliminating the need for an expensive oxygen plant when oxygen is used as gasification agent. This mitigates the diluting effect of N₂ from air and produces syngas with a higher heating value [2,16–19].

Pyrolysis has been confirmed to be the capital step of biomass thermochemical conversion and also a process of degradation using pyrolytic and catalytic mechanisms [2,11,18–20]. Han et al. summarized the mechanism of action of the catalyst on tar reforming in the following series of steps

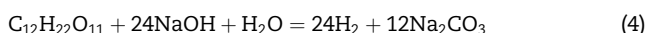
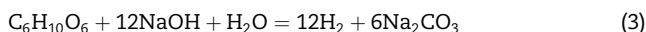
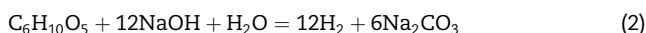
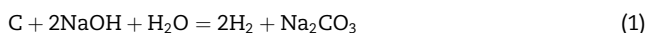
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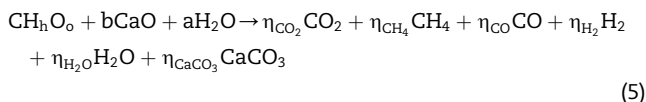
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[21]. First, methane or other hydrocarbons are dissociated and adsorbed onto a metal site where metal-catalyzed dehydrogenation takes place. Next, water hydroxylates the ceramic surface. At a specific temperature, the OH radicals migrate to the metal sites, leading to oxidation of the intermediate hydrocarbon fragments and surface carbon to form CO and H₂. Ishida et al. considered the gasification and adsorption reactions as follows [22,23]:



The overall reaction for steam gasification of biomass in presence of CaO can be written as follows [24–26]:



Despite of a great deal of research, however, biomass is still not the alternative of choice to fossil fuels for hydrogen and syngas production. The causes include discontinuous supplies of a fuel of limited resources and varying composition resulting in poor economy of small-scale systems and operating problems related to tars formation and corrosion, especially when the use of biomass is considered [3,27,28].

In a previous study, ionic catalysis was found to have significant effects on the formation of H₂ during the steam gasification process of cellulose. However, the mechanism by which C, H, and O elements are converted is not entirely clear [29]. It is assumed that the large-scale use of biomass in the field of hydrogen and syngas production must be essentially impeded by the unclear gasification mechanism. In this work, the conversion mechanism of C, H, and O elements is further supported by experiments, and the catalytic steam gasification of biomass is revealed.

Experimental procedure

A uniform mixture of cellulose (1.6214 g, based on (C₆H₁₀O₅) mass) and water (3.0000 g) was uniformly mixed to produce mixture M1. The mass ratio of sodium aluminum oxide (Al₂O₃·Na₂O) to sodium aluminum oxide and cellulose was set to 0.4:1. Sodium aluminum oxide (1.0809 g) and water (3.0000 g) were uniformly mixed, heated until boiling, and cooled to room temperature. Cellulose (1.6214 g) was further added and mixed uniformly to produce mixture M2. The mixtures M3 and M4 were prepared in correspondence to the mass ratio 0.5:1 and 0.6:1 of sodium aluminum oxide (Al₂O₃·Na₂O) to sodium aluminum oxide and cellulose respectively. NaOH (0.1200 mol), water (3.0000 g) and cellulose (0.0100 mol) were uniformly mixed to produce mixture M5. The mixtures M1–M5 were dried at 378 K (±1 K), crushed, and sieved to obtain the corresponding gasification samples S1–S5. The samples S1–S5 had a particle size of about

0.300–0.150 mm. The mass of cellulose in each sample (S1–S5) was approximately 0.3243 g (0.0020 mol).

Samples S1–S5 and their corresponding blank tests were evaluated under atmospheric pressure in a cylindrical stainless steel reactor. The air in the reactor was flushed out with Argon (Ar) before the test. The temperature was raised linearly from room temperature to 1073 K at a rate of 2.5 K/min. When the temperature reached 453 K, a flow of steam (V_{H₂O(g)} = 500 ml/min, 101,325 Pa, 373 K) carried by Ar (V_{Ar} = 100 ml/min, 101,325 Pa, 293 K) was passed through the reactor. The gaseous products were cooled using an ice–water mixture first followed by an ice–salt mixture. The analysis of the gaseous products was performed on-line using a gas chromatograph coupled with TCD and FID. The limit of detection of CO in the effluent gas was less than 1 × 10^{−6} mol/mol in the present study.

Results and discussion

The rates of formation of product gases at 473–1073 K of sample S1 were shown in Fig. 1 as a function of temperature. The rates of formation of CO and CO₂ at low temperatures were much higher than those of H₂ and CH₄. At low temperatures, CO and CO₂ were mainly produced from the inner reaction of carbon with oxygen in the cellulose. At high temperatures, the product gases of S1 were composed of H₂, CO, and CO₂, which came mainly from the water–gas reaction and the water–gas shift reaction [11,19,29].

The formation rates of the product gases of samples S2–S4 at 473–1073 K as a function of temperature were shown in Figs. 2–4, respectively. The rates of formation of CO and CO₂ from S2–S4 at low temperatures were much higher than those of H₂ and CH₄. At low temperatures, CO and CO₂ were mainly from the inner reaction of carbon with oxygen contained in cellulose. The product gases of S2–S4 at higher temperatures were mainly composed of H₂, CO, and CO₂, which came mainly from the water–gas reaction and the water–gas shift reaction, respectively [11,19,29].

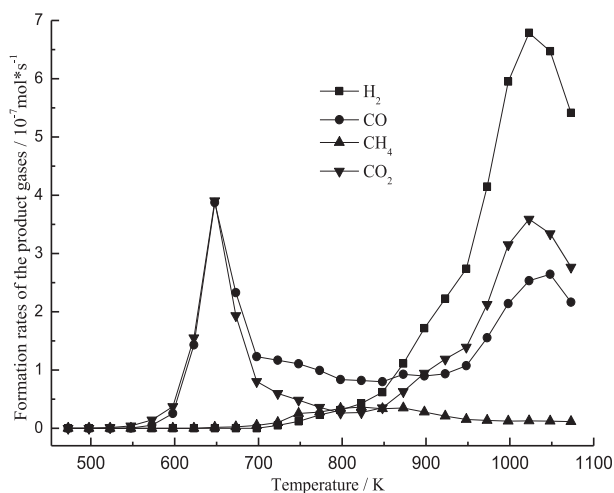


Fig. 1 – The formation rates of the product gases of the sample S1.

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