



## Hydrogen and syngas production by catalytic biomass gasification



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### ABSTRACT

Air-steam gasification of wood residue was explored in a research scale fluidized bed. Catalytic activity of two different kinds of metal catalysts (Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>) with various catalyst loadings (20, 30, and 40%) was also investigated at various residence time (20, 40, and 60 min) and gasification temperature (750, 825, and 900 °C). Non-catalytic experiments were also carried out to determine the optimum conditions for tar cracking and hydrogen/syngas production. Results were revealed that the high temperature (~900 °C) and high catalyst loading (~40%) are favorable for tar cracking and high-purity hydrogen production. It was also found that for a residence time of 60 min, the tar cracking at the presence of Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> is 196% more than that of the case without any catalyst, while at the presence of Ni/Al<sub>2</sub>O<sub>3</sub> it drops to 162%. Finally, the experiments were showed that Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> is more suitable for biomass conversion and hydrogen production than Ni/Al<sub>2</sub>O<sub>3</sub>.

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### 1. Introduction

Among all renewable energy sources such as wind, solar, and biomass energy, the latter appears promising for economic development specifically in small size power plants, located close to forestry regions where biofuels are plenty [1]. The use of biomass as one of the most important renewable energy sources may also decrease the emission of greenhouse gases and air pollutants.

The gasification of biomass that results in hydrogen and syngas, required for power generation and chemical compounds, poses the most promising effort to reduce the demand for fossil fuels, such as coal and oil, significantly [2]. Gasification is a thermochemical process that devolatilizes solid (coal, biomass, and municipal solid waste) or liquids (waste lubricant oil and olive oil) fuels, converting them into a high caloric value syngas, used as an energy carrier for power generation devices such as gas turbines and gas engines. Produced syngas also can be used an efficient feedstock to produce bio-diesel or converted to methane gas via the Sabatier, dimethyl ether (DME), and synthesis fuel via the Fischer-Tropsch process [3]. This is so since hydrogen can be used either directly as a feedstock in the Proton Exchange Membrane (PEM) fuel cells or as a

means to produce a large number of chemicals, such as methanol. There are four main solutions, possible for hydrogen production: gasification, water hydrolysis, biological processes, and hydrogen production by nuclear power with the latter, having important potential advantages compared to the other methods [4]; however, more researches need to be done due to technical problems such as safety requirements and nuclear wastes management. Although the biological and hydrolysis processes are the two main environmentally-friendly processes to produce a hydrogen-rich gas, they are not popular as a result of their low efficiency and high cost [5]. Among all methods for hydrogen production, gasification is the most interesting one, able to produce not only hydrogen but syngas. Here, the steam is used as a gasifying agent [6]. From a literature review, it was found that the gasification process may be done in presence of various gasification agents such as pure oxygen, steam, CO<sub>2</sub>, air or a mixture of oxygen and steam. Schuster et al. [7] concluded that the gasification by using air generates a synthesis gas with a heating value of 4–7 MJ/N m<sup>3</sup>, where gasification with a mixture of oxygen and steam produces the syngas having a heating value between 10 and 18 MJ/N m<sup>3</sup>.

One of the most important challenges during the steam gasification of biomass is the formation of tar which creates severe operational problems either in the gas engines for power generation or in the gasifier pipelines for syngas production [8]. There are several chemical and physical methods to remove or convert the tar

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such as catalytic hydrocracking (primary method) and gas cleaning system (secondary method).

A brief look at the literature review, related to the current research, shows a strong interest in tar conversion by means of metal-based catalysts. Asadullah et al. [9] explored the gasification of cedar wood using highly-efficient Rh/CeO<sub>2</sub>/SiO<sub>2</sub> catalysts in a continuous feeding fluidized bed reactor. Norouzi et al. [10] studied the hydrothermal gasification of *Enteromorpha intestinalis* (algal biomass) in a batch type reactor to evaluate its potential for hydrogen production in presence of Ru promoted Fe-Ni/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> nanocatalysts. Kang et al. [11] evaluated the potential of several catalysts and identified K<sub>2</sub>CO<sub>3</sub> and 20Ni-0.36Ce/Al<sub>2</sub>O<sub>3</sub> as best catalysts. They also found that the high temperature (~650 °C), and high catalyst loading (~100%) lead to higher hydrogen production.

From the above discussion, it can be easily discovered that only a few research papers have addressed the production of hydrogen-rich gases through catalytic gasification of biomass. Hence, there is a need for a catalytic study accordingly. In our previous work, we used the gasification technology to produce hydrogen gas without catalyst loading, figuring out that the level of tar is much higher than the standard level for engineering applications [12]. In this research work, however, for the first time to our knowledge, a research scale fluidized bed is used to study the catalytic gasification of wood residue with various Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> loading for the production of hydrogen rich gas.

## 2. Materials and methods

The setup and the experimental method have been described in details before [12]. Hence, only a brief description is given here. The experimental tests have been carried out in an experimental rig, including a research scale fluidized bed (3.5 m tall  $\times$  0.3 m diameter) a cooling system, a fuel feeding system which allows a controlled fuel flow be supplied, and a fuel storage tank (10.2 m<sup>3</sup>). The gasification agent is a mixture of oxygen and steam, the flow rate of which is controlled by means of a water pump placed upstream of the steam generator. The catalysts, used in this research, are Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> with  $1.3 \times 10^{-4}$  mol Ni/g of catalyst and various contents of CeO<sub>2</sub>, prepared via the sequential impregnation method. To change the Al<sub>2</sub>O<sub>3</sub> from  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> to  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub> have been calcined in air at 773 K for 2 h before reaction. Also Ni/CeO<sub>2</sub> have been prepared by impregnating CeO<sub>2</sub> using the aqueous solution of Ni (NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and SiO<sub>2</sub> (Aerosil, 325 m<sup>2</sup>/g). The loading of CeO<sub>2</sub> on Al<sub>2</sub>O<sub>3</sub> was in the range of 20–40 mass%. After the Ce salt was loaded on Al<sub>2</sub>O<sub>3</sub>, it was dried at 375 K for 10 h and then it was calcined at 785 K for 2.5 h under atmospheric condition. The all catalyst particles were crushed and sieved to flours with the size of 35  $\mu$ m.

## 3. Results and discussion

We have prepared two different types of catalysts (Ni/Al<sub>2</sub>O<sub>3</sub> and Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>) with different Ni loadings and examined them in the gasification of wood residue in a research scale fluidized-bed gasifier. At first, the feedstock thermally decomposed to the volatiles, char, and small fraction of gaseous products. Then the tar contents and char particles reacted with the Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> and Ni/Al<sub>2</sub>O<sub>3</sub> particles in the lower dense region where steam and oxygen are present and a part of the volatiles and solids may take part in the firing and oxidation reactions to form greenhouse gases. Table 1 gives the performance of the catalysts with respect to the syngas yield, gas composition, char conversion, tar yield, and cold gas efficiency. In general, the formation rate of the tar and char is a function of the operating conditions (Equivalence Ratio (ER), gasification agent, system pressure, and temperature), hydrody-

namic characteristics of the bed (fluidized, fixed, or moving bed), and the type of feedstock (coal, municipal solid wastes, biomass, or heavy oil). Another important factor is the nature of the catalyst used which improves the quality of the produced syngas, converting the tar content and unburned char to gas products. Compared to Ni/Al<sub>2</sub>O<sub>3</sub> catalysts, it has been found that the performance of Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> catalysts is more effective for increasing the syngas yield and decreasing that of tar and char. Another possible solution to decompose the tar yield is when the temperature enjoys a significant ascent; however, it is well known that an increase in the temperature may cause the catalyst deactivation, not to mention a series of structural problems, related to the destruction of the catalysts structure [13]. As mentioned above, the addition of catalyst also affects the composition of the produced syngas and char yield. Since the addition of catalyst enhances the gas products, it might increase the reformation of light tars inside the gasifier, advancing carbon conversion in turn [14]. Gasification efficiency (cold gas basis) is an important factor to evaluate the performance of the system. It is defined as the ratio of chemical energy of the syngas, represented by higher heating value of the produced syngas, to the higher heating value of the feedstock. Since the ratio is very sensitive to ER, it is necessary to maintain ER at a permissible level so that the syngas production can be sustained. More significantly, with Ni/Al<sub>2</sub>O<sub>3</sub>/CeO<sub>2</sub> a high caloric value syngas is obtained with a negligible formation of tar and unburned char. As we have seen, these data confirm the good catalytic performance of Ni/Al<sub>2</sub>O<sub>3</sub>/CeO<sub>2</sub>.

Fig. 1 shows the tar yield, obtained at different temperatures and with varied catalysts. Tar plays an important role in the process as it results in serious operational problems and blocks the pipelines. As illustrated in Fig. 2, higher temperatures significantly resulted in tar cracking. Based on the Le Chatelier's principle, higher temperatures favor the products in endothermic reactions. Methane reformation and tar cracking are endothermic, hence becoming of more important at higher temperatures [15]. If the temperature increases from 750 to 900 °C, the yield of tar reduces while the syngas yield gets enhanced (Table 1). At the presence of steam, Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> is active to enhance the syngas yield and lower the rate of tar formation. Results also show that there is a strong potential for tar cracking by the catalysts, even though the catalytic activity of Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> for the reformation of tars and char (not shown here) is more compared with that of Ni/Al<sub>2</sub>O<sub>3</sub>. As aforementioned, the possible reason for this phenomenon is the improvement of the surface area of Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> due to the sintering of CeO<sub>2</sub> during the process. It seems that the influence of catalysts on tar yield is more significant at higher temperatures. According to the data, reported in the literature, the increase of

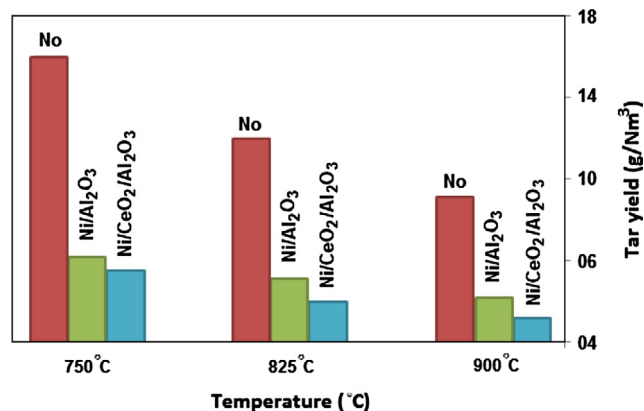


Fig. 1. Influence of temperature and catalyst on tar yield. (ER = 0.17, steam/biomass = 0.6 (wt/wt).)

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