



Fast microwave-assisted catalytic gasification of biomass for syngas production and tar removal



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HIGHLIGHTS

- We developed a microwave-assisted biomass catalytic gasification system.
- Ni/Al₂O₃ was the most effective catalyst and the gas yield reached above 80%.
- Ni/Al₂O₃ had good stability against deactivation during gasification process.
- We put forward a new concept of microwave-assisted dual fluidized bed gasifier.

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ABSTRACT

In the present study, a microwave-assisted biomass gasification system was developed for syngas production. Three catalysts including Fe, Co and Ni with Al₂O₃ support were examined and compared for their effects on syngas production and tar removal. Experimental results showed that microwave is an effective heating method for biomass gasification. Ni/Al₂O₃ was found to be the most effective catalyst for syngas production and tar removal. The gas yield reached above 80% and the composition of tar was the simplest when Ni/Al₂O₃ catalyst was used. The optimal ratio of catalyst to biomass was determined to be 1:5–1:3. The addition of steam was found to be able to improve the gas production and syngas quality. Results of XRD analyses demonstrated that Ni/Al₂O₃ catalyst has good stability during gasification process. Finally, a new concept of microwave-assisted dual fluidized bed gasifier was put forward for the first time in this study.

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1. Introduction

Currently, increasing researches have been conducted on sustainable energy sources, as an alternative to traditional fossil fuels. Since biomass is a carbon-neutral energy source (Mckendry, 2002), the efficient uses of biomass are considered very promising in the future energy portfolio (Richardson et al., 2012). Among all the utilization technologies, the production of syngas from biomass gasification is considered as an attractive route to produce chemicals, biofuels, hydrogen and electricity (Damartzis and Zabaniotou, 2011; Kirkels and Verbong, 2011; Lin and Huber, 2009). It has been estimated that syngas production from biomass accounts for at least half, and in many cases more than 75% of the cost of biofuel

production (Hamelinck and Faaij, 2002; Spath and Dayton, 2003). Therefore, the successful development of cost-effective processes for high-quality syngas production will greatly promote biomass utilization.

The gas produced from biomass gasification is mainly composed of H₂, CO, CO₂, CH₄ and some light hydrocarbons and also contains contaminants such as H₂S, HCl, tar, and solid particles. Among all the contaminants, tar is the most common and troublesome compound and has been extensively discussed in previous studies (Anis and Zainal, 2011; Li and Suzuki, 2009; Torres et al., 2007). Tar is a complex mixture of organic chemicals largely composed of aromatic hydrocarbons and can cause serious problems including fouling of engines and deactivation of catalysts, due to its condensation and polymerization (Devi et al., 2003, 2005; Han and Kim, 2008). Therefore, some strategies such as catalytic gasification have been considered to reduce tar content in syngas.

Traditional types of biomass gasification reactors include fixed bed and fluidized bed (Dong et al., 2010; van der Meijden et al.,

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2009; Xie et al., 2012). Dong et al. (2010) studied gasification of coffee grounds using a two-stage dual fluidized bed gasifier (DFBG), obtaining syngas with H₂ and CO contents of 31.23% and 29.20%, respectively. The tar content was reduced from about 40 g/m³ to 10 g/m³ raw gas with Ca impregnation onto fuel. Employing a circulating fluidized bed (CFB) as the gasifier and a bubbling fluidized bed (BFB) as the combustor, (van der Meijden et al. (2009)) examined steam gasification of wood pellets and obtained syngas containing around 38% of H₂, 19% of CO and 40 g/m³ of tar at the temperature of 925 °C. Xie et al. (2012) investigated two-stage catalytic pyrolysis and gasification of pine sawdust in a fixed bed reactor and observed a maximum syngas yield of 3.29 m³/kg biomass. However, the yield of liquid fraction was around 15–20 wt% of dry biomass.

Microwave irradiation is an alternative heating method and has already been successfully applied to biomass pyrolysis (Bu et al., 2012; Du et al., 2011; Wang et al., 2012). Compared with conventional heating processes where heat is transferred from the surface to the core of the material through conduction driven by temperature gradients, microwaves induce heat at the molecular level by direct conversion of the electromagnetic energy into heat (Sobhy and Chaouki, 2010), and therefore, they can provide uniform internal heating for material particles. In addition, the instantaneous response of microwave makes it easier for a rapid start-up and shut-down. Furthermore, the process operation involves a simple set-up and can be easily adapted to currently available large-scale industrial technologies. Microwave heating is a mature technology and development of microwave heating system is of low cost. Although many advantages of microwave heating over traditional heating methods and some progress made in biomass pyrolysis, no research has been conducted in biomass gasification using microwave technology.

In this study, microwave-assisted gasification of biomass was carried out under different conditions. Catalysts including Fe, Co and Ni with Al₂O₃ support were selected and compared for their effects on syngas production and tar removal. X-ray diffraction (XRD) analyses of catalysts before and after reactions were conducted to study their stability during gasification process. In addition, the effect of steam on syngas yield and quality was also investigated.

2. Methods

2.1. Materials and catalysts

The corn stover chosen as the biomass material for this study was obtained from a farm field located in Saint Paul Campus, University of Minnesota (Twin Cities). The basic physico-chemical characteristics of the corn stover including proximate analysis and element analysis were conducted. The moisture content of corn stover is 5.3% on wet basis and the volatile content is 81.9%. According to the elemental analysis, the simplified chemical formula of the raw material that derives is CH_{1.53}O_{0.97}. In addition, the higher heating value (HHV) and net heating value (NHV) calculated using equations (Vallios et al., 2009) are 15.1 MJ/kg and 13.1 MJ/kg, respectively. Prior to its use, the corn stover samples were ground using a rotary cutting mill and then screened to limit the particle size smaller than 0.5 mm. Afterwards, these ground samples were dried for more than 24 h at 80±1 °C.

The catalysts used in the experiments included Fe/Al₂O₃, Co/Al₂O₃, and Ni/Al₂O₃ prepared by impregnating porous alumina (60 mesh, surface area 150 m²/g) in nitrate solution. Alumina was used as the catalyst support. Catalyst loading of 15% was used for all the three catalysts. After impregnation for 12 h, the catalysts were dried at 105 °C, and then ground and screened to achieve a

particle size smaller than 3 mm. After being calcined at 500 °C in a muffle furnace for 4 h, Fe-, Co- and Ni-based catalysts were reduced at 500 °C, 350 °C and 450 °C, respectively, using a gas mixture of H₂/He (200 sccm) with a molar ratio of 1:1 for 12 h prior to application.

2.2. Apparatus

The tests of biomass catalytic gasification were performed in a microwave oven (MAX, CEM Corporation), with the power of 750 W at a frequency of 2450 MHz. The microwave-assisted biomass catalytic gasification system is composed of a biomass feeder, a microwave oven, a quartz reactor with a layer of microwave absorbent bed inside, thermocouples (K-type) to measure the temperatures of oven cavity and bed particles, condensers and liquid fraction collectors, a gas collector, and some quartz connectors. For safety purpose, a microwave detector (MD-2000, Digital Read-out) was used to monitor microwave leakage.

First of all, 800 g of SiC particles with particle size of 30-grit were put in the quartz reactor as the microwave absorbent bed. The reactor was then placed in the cavity of the microwave oven. After the inlet and outlet quartz tubes connected with the reactor, the microwave oven was turned on for heating process. For each experiment, the sample was prepared by physically mixing 15 g corn stover with 5 g catalyst. When the temperature of bed particles reached the set temperature of 900 °C, the prepared sample was dropped through the feeder onto the hot SiC bed, meanwhile the microwave oven was controlled to be on or off in order to keep the temperature of absorbent bed stable. Flowing through the condensers, the gas product was collected into sampling bags for offline analysis, with tar condensed into the liquid collectors for subsequent analysis. The yields of solid and liquid fractions were calculated on the basis of their own weight, while the gas yield was calculated by difference based on the mass balance.

2.3. Gas and tar analysis

Offline gas analysis was performed using a Varian CP4900 Micro-gas chromatograph (GC) with a thermal conductivity detector (TCD). The two columns used were PoraPlot Q and 5 Å molecular sieve with helium as carrier gas. The temperatures of both injector and detector were set at 110 °C. The temperatures of PoraPlot Q and 5 Å molecular sieve columns were kept at 80 °C and 150 °C, respectively.

The components of liquid product were specified using an Agilent 7890-5975C gas chromatography/mass spectrometer (GC/MS) with a HP-5 MS capillary column. Helium was employed as the carrier gas at a flow rate of 1.2 mL/min. The injection size was 1 µL with a split ratio of 1:10. The initial oven temperature was 40 °C held for 3 min and then increased to 290 °C at a rate of 5 °C/min, and held at 290 °C for 5 min, while the injector and detector were maintained at constant temperature of 250 °C and 230 °C, respectively. The compounds were identified by comparing their mass spectra with those from the National Institute of Standards and Technology (NIST) mass spectral data library.

2.4. Catalyst characterization

The X-ray powder diffraction (XRD) patterns, obtained on a Siemens D5005 X-ray diffractometer instrument with a Cu Kα radiation at 45 kV and 40 mA, were used to identify the major phases present in the catalysts.

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