



Hydrogen production from banyan leaves using an atmospheric-pressure microwave plasma reactor



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HIGHLIGHTS

- Pyrolysis of banyan leaves is studied in a microwave plasma reactor.
- Increase the microwave power levels results in an increase of H₂ production.
- H₂ production rate is 20.44 mg min⁻¹ at 1000 W.
- H₂ production efficiency is 67.33% at 1000 W.

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ABSTRACT

Growth of the hydrogen market has motivated increased study of hydrogen production. Understanding how biomass is converted to hydrogen gas can help in evaluating opportunities for reducing the environmental impact of petroleum-based fuels. The microwave power used in the reaction is found to be proportional to the rate of production of hydrogen gas, mass of hydrogen gas produced per gram of banyan leaves consumed, and amount of hydrogen gas formed with respect to the H-atom content of banyan leaves decomposed. Increase the microwave power levels results in an increase of H₂ and decrease of CO₂ concentrations in the gaseous products. This finding may possibly be ascribed to the water–gas shift reaction. These results will help to expand our knowledge concerning banyan leaves and hydrogen yield on the basis of microwave-assisted pyrolysis, which will improve the design of hydrogen production technologies.

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1. Introduction

Recently, researches pay more attention on global warming and energy issues. The usage of fossil fuels increased lot of greenhouse gases. Those greenhouse gases caused global warming effects. Therefore, how to reduce the greenhouse gas emissions became the special issue. Many studies focused on alternative energy such as solar energy, wind energy, tide energy, geothermal energy, biomass energy and so on. In recent years, energy from waste has become a possible solution to reduce wastes and produce clean energy. Biomass conversion technologies include biological (anaerobic/aerobic digestion and fermentation) and thermal methods. Pyrolysis is one of the most promising thermo-chemical conversion technologies for recovering energy from biomass. Pyrolysis converts biomass to gas (hydrogen and syngas), liquid (bio-oil,

methanol and ethanol), and solid (coal). The gases produced by pyrolysis are mainly H₂, CH₄, CO and CO₂ (Mountouris et al., 2006; Huang and Tang, 2007; Huang et al., 2008, 2010). The thermal conversion of biomass to gas is a possible option because the syngas can be easily stored and transported (Wu and Williams, 2009; Buffoni et al., 2009; Efiika et al., 2012; Xie et al., 2012).

Some studies focused on pyrolysis by using microwaves as heating resource to convert biomass (Chen et al., 2008; Lei et al., 2009; Bu et al., 2012). The major gas products from rice straw under microwave pyrolysis were H₂, CO₂, CO, CH₄ at concentrations of 55, 17, 13, 10 vol%, respectively (Huang et al., 2008, 2010). Microwave pyrolysis of coffee hulls, waste tea, corn stover, wheat straw, and oil palm biomass led to high H₂ content gas products, implying that microwave-induced pyrolysis of biomass has the potential to produce H₂-rich fuel gas (Lei et al., 2009). Hydrogen is a generally acknowledged renewable, recyclable and clean fuel. Hydrogen does not content carbon when compared to hydrocarbon fuel (Dominguez et al., 2007; Yagmur et al., 2008; Wan et al., 2009; Budarin et al., 2009; Salema and Ani, 2011). Hydrogen has higher

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flame speed, wider flammability limits, and faster burning velocity than diesel fuel (Bari and Esmaeil, 2010).

Recently, an innovative plasma gasification technology was developed and it seems to be the most effective and environmentally friendly method for biomass treatment and energy utilization (Mountouris et al., 2006; Huang and Tang, 2007; Lemmens et al., 2007; Bratsev et al., 2006). Plasma systems have been used to create gasification conditions and produce synthetic gases (syngas) from biomass under high temperatures (typically >1000 °C) (Reed et al., 1981). Plasma system ionised gas to create substantial temperatures that can dissociate biomass into its elemental constituents. Microwave plasma is an established technology and has been used for a variety of applications such as biomass gasification, chemical vapor deposition, and bacterial sterilisation (Reed et al., 1981; Fünér et al., 1995; Purevdorj et al., 2002; Matusiewicz, 2002). Although there has been a lot of researches focused on microwave pyrolysis and plasma systems for biomass gasification, the topic of using microwave plasma to produce hydrogen from biomass wastes, banyan leaves, has seldom been addressed. Biomass wastes, banyan leaves, played a role in future renewable energy generation. This study investigated hydrogen and other gases from biomass wastes, banyan leaves (*Ficus microcarpa*), fed into the microwave plasma system. The resultant gas from the process was analyzed in real-time using a gas analyser. The effects of plasma power are investigated. Finally, hydrogen production rate (mg min^{-1}), hydrogen production factor ($\text{mg-H}_2 \text{ g-biomass}^{-1}$) and conversion rate (%) were calculated and evaluated.

2. Experimental section

2.1. Methods

The hydrogen production rate R_{H_2} (mg min^{-1}) is determined by:

$$R_{\text{H}_2} = C_{\text{H}_2} \times Q_{\text{gas}} \times \frac{1 \text{ mole}}{24.5 \text{ L}} \times MW_{\text{H}_2}$$

where C_{H_2} is the hydrogen volume contents (vol%), Q_{gas} is flow gas rate (12 L min^{-1}), MW_{H_2} is molecular weight of hydrogen, and 24.5 L is the ideal-gas volume for one mole at temperature of 298 K.

2.2. Experimental setup

Fig. 1 shows the experimental setup comprising the electrodeless microwave excited atmospheric plasma system (APS) and the product analysis system. Details of this experimental setup are described in references (Tsai and Chen, 2009; Wang et al., 2010a, 2010b). Experiments in the present study are repeated at least three times to provide evidence of reproducibility.

2.3. Pyrolysis procedure

The pyrolysis experiments are conducted in an atmospheric-pressure microwave plasma reactor at applied microwave power of 800, 900 and 1000 W corresponding to the temperature of 1063, 1093 and 1121 K in the plasma zone. Approximately 1 g per 5 s of dry banyan leaves at room temperature is fed with axial flow into the reacting zone from upstream of the cavity resonator and N_2 is used as bath gas at a flow rate of 12 L min^{-1} (axial flow rate of 3 L min^{-1} and swirl flow rate of 9 L min^{-1}). The flow rate of N_2 , supplied from compressed gas tanks, is kept constant. The reactor is made from a quartz tube of 35 cm length and an inner and outer, diameter of 2.9 and 3.3 cm, respectively.

2.4. Banyan leaves and gas analysis

Both the pyrolysis characteristics and product distribution of banyan leaves are analyzed in this study. Morphological changes of the banyan leaves samples before and after pyrolysis are observed by an environmental scanning electron microscopy (ESEM). Elemental chemical analysis (C, H, N, S and O) of banyan leaves and the residue after pyrolysis are performed on an Elementar Vario Micro Cube elemental analyzer (EA). Sampling is accomplished by continuously withdrawing gases from within the plasma zone using a micro probe with a capillary probe of 0.1 mm i.d. A gas chromatography (GC), equipped with capillary columns (type: Restek™ 13,821, length: 15 m, I.D. 0.32 mm, membrane thickness: 0.25 μm , stationary phase: DB-35ms, MR2, VF-35ms), a thermal conductivity detector (TCD) and a residual gas analyzer (RGA), are used for stable species measurements. The RGA is used to monitor the hydrogen concentration produced over the entire pyrolysis history. For the H_2 analyzed in GC/TCD, the carrier gas is nitrogen and the detector oven and vaporizer temperatures are 513 and 383 K respectively. The GC oven temperature is set to 383 K for 10 min and ramped to 473 K at 288 K min^{-1} held for 14 min. For the CO and CO_2 analyzed in GC/TCD, the carrier gas is helium and the detector oven and vaporizer temperatures are 393 and 323 K respectively. The GC oven temperature is set to 333 K for 5 min and ramped to 498 K at 293 K min^{-1} held for 10 min.

3. Results and discussion

3.1. Element analysis and ESEM

In this study, produced syngas includes hydrogen rich gas through the dry banyan leaves pyrolysis reaction in the microwave plasma processing at different levels of absorbed microwave power. Fresh samples were washed four times with filtered water and sun-dried for four days. The cleaned dry banyan leaves were then finely chopped into pieces and then ground into fine powder. The element analysis of dry banyan leaves before and after pyrolysis was shown in Table 1. The contents of C, H, O, N, and S of dry banyan leaves before pyrolysis were 44.79, 6.66, 43.15, 1.46, and ND wt%, respectively. In other words, the mole ratio of C:H:O was 6:10.68:4.32. The ratio of C, H and O atoms suggests the presence of carbohydrate isomer to a cellulose monomer with chemical structure of $\text{C}_6\text{H}_{10}\text{O}_5$, and hence implies that this biomass is composed mainly of cellulose. The contents of C, H, O, N, and S of dry banyan leaves after pyrolysis were 54.01, 1.76, 19.88, 0.60 wt%, and ND, respectively. In other words, the mole ratio of C:H:O was 1:0.79:0.21. Since the coke formation after microwave plasma pyrolysis, the weight ratio of carbon element increases after microwave plasma reaction. Similar results were found by ESEM. The ESEM was shown in Fig. S1. It was also found that the surface structure of dry banyan leaves is cracked and become fragmented during attempts to thermally decompose the solid residue. The above results displayed pyrolysis of dry banyan leaves happened using microwave plasma system.

3.2. Time-history measurements for H_2 formation

A general profile of reactions product H_2 as measured by the RGA as function of time during the pyrolysis is displayed in Fig. 2. Reactions are observed to occur rapidly, with product evolution detected on residual gas analyzer immediately after feeding. At the first stage of the pyrolysis, where $t < 20 \text{ s}$, there is a steep increase of H_2 production ($10 < t < 20 \text{ s}$) after short induction time ($0 < t < 10 \text{ s}$). This stage, however, has only about one-third of hydrogen molecules formed. It is worth to note that the needed

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