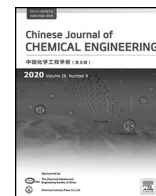




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Article

Solid oxide fuel cells in combination with biomass gasification for electric power generation

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ABSTRACT

Biomass, a source of renewable energy, represents an effective substitute to fossil fuels. Gasification is a process that organics are thermochemically converted into valuable gaseous products (e.g. biogas). In this work, the catalytic test demonstrated that the biogas produced from biomass gasification mainly consists of H₂, CH₄, CO, and CO₂, which were then be used as the fuel for solid oxide fuel cell (SOFC). Planar SOFCs were fabricated and adopted. The steam reforming of biogas was carried out at the anode of a SOFC to obtain a hydrogen-rich fuel. The performance of the SOFCs operating on generated biogas was investigated by I–V polarization and electrochemical impedance spectra characterizations. An excellent cell performance was obtained, for example, the peak power density of the SOFC reached 1391 mW·cm^{−2} at 750 °C when the generated biogas was used as the fuel. Furthermore, the SOFC fuelled by simulated biogas delivered a very stable operation.

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1. Introduction

Although the massive consumption of fossil fuels has brought about greenhouse effects and environmental pollution problems, they are still the most important source of energy over several decades. Because the reserves of fossil fuels are very limited, and likely to be exhausted within 50 to 100 years, we need to find alternative low-pollution fossil fuels and renewable energy sources. With the support of government policies, renewable energy sources for example solar, wind, hydraulic and biomass have attracted more and more attention in the past 20 years. Among these renewable energy sources, the use of biomass has a very long history, dating back to the earliest uses of energy by humans [1,2]. When it is used in a planned way, the biomass energy has a renewable nature. Biomass energy is widely distributed and can be quickly produced. When biomass energy is used less quickly than it is being produced, we assume that there are no CO₂ emissions in the process, or that CO₂ is cycling. Additionally, residues and wastes of the biomass feedstock produce inappreciable greenhouse gas emissions, being part of a short carbon cycle. It is estimated that bioenergy including biomass and wastes will contribute 25%–35% of global prime energy supply in 2050 [3,4].

However, the large-scale utilization of biomass energy is still confronted with some problems. The collection of biomass requires a lot of manpower and material resources due to its wide distribution, which makes the transportation cost of biomass higher. If biomass is vaporized and biomass gas (i.e. biogas) is obtained, the cost of energy transmission can be reduced [5–7]. There are many biomass gasification methods, including the fermentation by biological methods, the direct pyrolysis, and the reforming with air, oxygen or steam at high temperatures [8–12]. Among these methods, the biomass reforming gasification, especially the high-temperature steam reforming, which produces the biogas with a high calorific value, has attracted the most attention because of its high efficiency and low environmental pollution. Biogas is generally composed of H₂, CO, CO₂, CH₄, and other gas components, can be applied to direct combustion power generation [13–15].

The energy conversion efficiency of thermal power plants is limited by the Carnot cycle, which also will reduce the utilization efficiency of biomass energy. To improve the efficiency of fuel power generation, fuel cells are a good choice [16–18]. Fuel cells are a kind of power generation device that directly transforms the chemical energy of their fuel into electric energy. The advantages of power generation using fuel cells are high efficiency and reduced environmental pollutants. Unlike conventional fuel cells, solid oxide fuel cells (SOFCs) operate at higher temperatures, with high catalytic performance and wide adaptability to different fuels [19–21]. In addition to hydrogen,

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hydrocarbon gases, liquids, and even solids can be used as the fuel for SOFCs [22–26].

An important aspect of biomass steam reforming is its ability to produce biomass gas as the fuel of SOFCs [27–30]. At present, more and more theoretical and experimental research studies have been carried out [31,32,34,35]. Campitelli *et al.* analyzed the integration of an SOFC operation with a biomass gasification process. The results show how a variation of the hydrogen utilization of SOFCs, the effects of which are correlated with the gasifier air requirement, affects electrical power output, hinging upon the moisture content of biomass [31]. Minutillo *et al.* have developed a small co-generation power plant fed by biomass and reported the performances. The power plant consisted of a micro gas turbine and an SOFC unit. They demonstrated that the hybrid configuration is very helpful to produce electric and thermal power at the same time from biomass with high efficiency [32].

There are mainly two types of SOFC in configurations based on shape: tubular and planar [33]. Chen *et al.* have carried out the gasification of mallee wood and wheat straw to fuel a tubular SOFC. Preliminary results have shown that the SOFC fueled by the biogas can reach a maximum power density of $576 \text{ mW} \cdot \text{cm}^{-2}$ at 800°C , comparable to one using an equivalent amount of pure H_2 or CH_4 [34]. Compared to tubular SOFCs, planar SOFCs have the advantages of larger volume energy density and greater ease of assembly. They have generated a lot of attention and been applied in demonstration industrial operations [35–38]. Gadsbøll *et al.* have conducted experimental studies on an 800 W planar SOFC stack with the real product gas originated from a two-stage gasifier as the fuel. The SOFC stack was operated at 700°C and 145 h of stable operation was reached without a significant decrease in SOFC performance [35].

Pine sawdust is a widely produced by-product of wood processing, and it could actually be put to good use if an efficient treatment system is available. For example, the production of bio-oil from the pine sawdust by pyrolysis is a typical treatment process [39–41]. Different from other biomass sources, pine sawdust is mainly obtained from the wood processing, so it is more convenient to collect and the cost is lower. It is a very promising development if we combine the pine sawdust gasification with the SOFC system to generate electricity by re-using. However, as far as we know, there are almost no related studies in the literatures. In this study, the high hydrogen-content gas was produced via air-steam gasification in a fixed bed, at high temperature, using pine sawdust biomass. The fuel gas was then introduced to an anode-supported planar SOFC as the fuel for an electrochemical performance test. The results demonstrated that a remarkable output was achieved by the SOFC.

2. Materials and Methods

2.1. Gasification of pine sawdust

The high-temperature steam gasification of pine sawdust was carried out in our lab with a fixed bed reactor. The temperature of the gasification was approximately 950°C . Air and water were introduced to the fixed bed with a compressor and pump during the gasification process. The air equivalence ratio was 0.2 and the steam to biomass ratio (S/B, in mass) was 0.8. The gasification product gases were cooled in ice water and collected in an air bag. The product gases in the air bag were introduced to a gas chromatograph (GC-9860, Hope, Nanjing, China) with a thermal conductivity detector (TCD) and flame ionization detector (FID). Because the amount of gas produced in our lab could not meet the needs of the SOFC, we also used a simulated biogas instead of the product gas derived from the biomass gasification. In this paper, biogas refers to the gas from the fixed bed reactor and simulated biogas refers to the gas mixture that has a similar composition to the biomass product gas.

2.2. Fabrication of anode-supported SOFC

An anode-supported SOFC was fabricated with tape-casting and spraying processes. Anode substrates, composed of commercial NiO (Chengdu Shudu Nano-science, China), and $(\text{Y}_2\text{O}_3)_{0.08}(\text{ZrO}_2)_{0.92}$ (YSZ, Tosoh), were prepared using a tape-casting process. To prepare the anode slurry for the tape-casting process, NiO, YSZ, starch (as a pore former), and fish oil (as a surfactant) were first ball-mill mixed at the mass ratio of 60:40:10:3 with a mixture of ethanol and dimethylbenzene for 24 h. Secondly, polyvinyl butyral (as a binder), polyethylene glycol (as a plasticizer), and dibutylphthalate (as a plasticizer) with the mass ratio of 14:10:13 were added to the slurry and, then, the slurry was ball-milled again for another 24 h. The slurry was cast onto a tape after vacuum pumping to remove the air, and the cast slurry was allowed to dry, in air, overnight. Finally, the anode substrates were detached from the tape and were then drilled to form disks with diameters of 16 mm. The disks were then calcined at 1100°C for 2 h to create sufficient mechanical strength. Next, a thin-film YSZ electrolyte layer was deposited onto each anode substrate via a wet powder spraying technique. The anode/electrolyte half-cells were then sintered at 1400°C for 5 h in air. In this work, $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}\text{-Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (BSCF-SDC, 7:3) was the cathode material. To avoid the solid reaction between YSZ and BSCF, a thin layer of SDC was deposited onto the surface of the YSZ electrolyte and then sintered at 1300°C for 5 h. Finally, the BSCF and SDC that were synthesized through the use of an EDTA-citric acid complexing sol-gel process were mixed with some organic solvent in a high-speed ball mill to produce the cathode slurry. The cathode slurry was sprayed over the central surface of the electrolyte, giving an effective area of 0.45 cm^2 , and then the cell was fired at 1000°C for 2 h in air.

2.3. Characterization

The catalytic activities of the anodes of the gasification product gas were tested in a flow-through reactor. All of the particles of the various anodes were reduced by H_2 at 700°C for 30 min and the temperature range of this reaction was from 600 to 800°C . The catalytic activities of the various anodes were studied in a flow-through type of fixed-bed, quartz-tube reactor with an inner diameter of $\sim 8 \text{ mm}$. 0.2 g of catalyst particles, having a size of 60 to 80 mesh, were placed in the middle of the reactor. A simulated gas, instead of the gasification product gas, was bubbled through water at approximately 25°C and fed into the reactors at a flow rate of simulated gas/helium = 20/80 $\text{ml} \cdot \text{min}^{-1}$ [STP] for reforming. The flow rate was controlled using AFC 80MD digital mass flow controllers (MFCs, Qualiflow). The gas mixtures were introduced from the top of the reactor; the effluent gases at the bottom of the reactor were introduced to a gas chromatograph (GC-9860, Hope, Nanjing, China) with TCD and FID to determine their compositions.

For the electrochemical performance test, the anode-supported SOFC was fixed onto a quartz tube (using silver paste) to form an anode chamber, which was then heated to 700°C at a rate of $2^\circ\text{C} \cdot \text{min}^{-1}$ and held for 2 h to set the seal. The cell was then heated to 750°C at a rate of $5^\circ\text{C} \cdot \text{min}^{-1}$ and hydrogen at a flow rate of $40 \text{ ml} \cdot \text{min}^{-1}$ (STP) was introduced into the anode chamber to begin *in situ* reduction of anodic NiO to metallic nickel. After approximately 2 h, the hydrogen flow rate was increased to $80 \text{ ml} \cdot \text{min}^{-1}$ (STP) and it was bubbled through water at room temperature. Fig. 1 presents the schematic diagram of the fuel supply system for the SOFC. I–V curves were collected at 50°C intervals over a temperature range of 600– 800°C using a digital electronic load (model IT8510, Itech, China) with a four-probe configuration. Electrochemical impedance spectroscopy (EIS) of the fuel cell under open circuit voltage (OCV) condition was undertaken and measurements were made with an electrochemical station (VersaSTAT 3, Ametek, the USA). EIS results were recorded in a frequency range of 100 kHz to 0.1 Hz with a signal amplitude of 10 mV. The biogas and simulated biogas were also

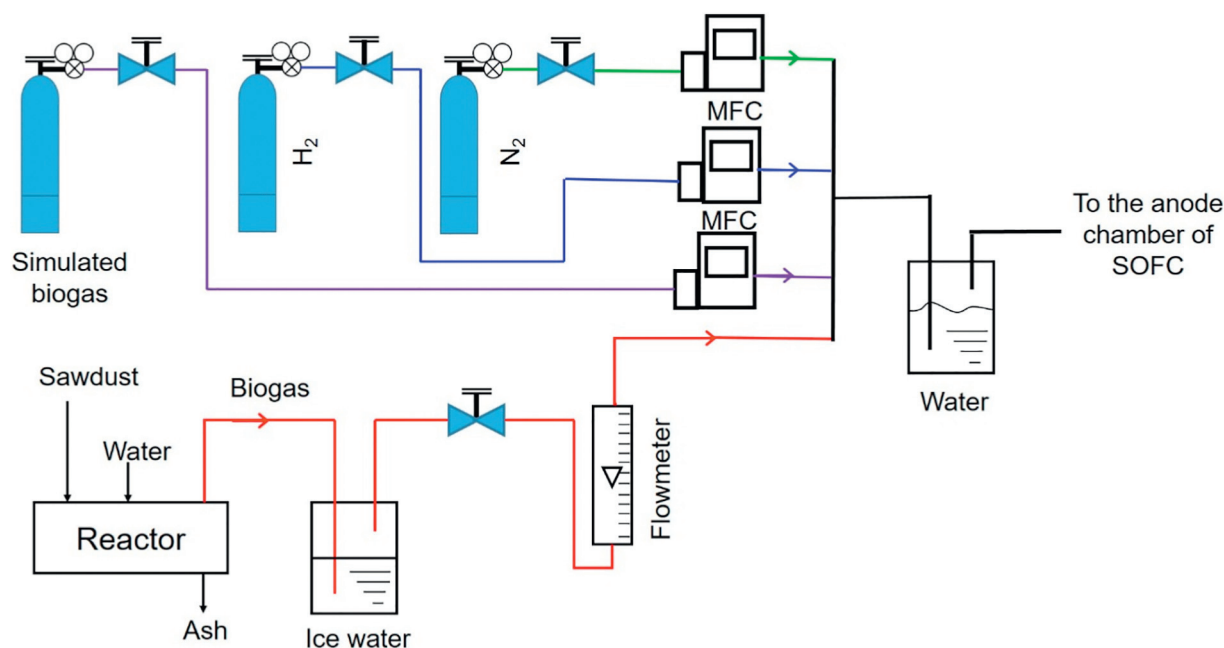


Fig. 1. Schematic diagram of the fuel supply system for the SOFC.

introduced to the anode of the cell for electrochemical performance testing at 750 °C. The flow rate of the biogas and simulated biogas was 80 ml min⁻¹. The flow rate of the fuel gases was regulated by the MFCs. After the test, the morphologies of the anode-supported SOFC were cut into small pieces and examined using a field-emission scanning electron microscope (FESEM, Hitachi S-4800, Japan).

3. Results and Discussion

3.1. Gasification of biomass

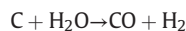
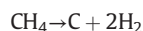
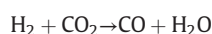
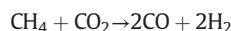
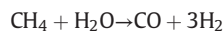
The raw material used for gasification in this work was pine sawdust, which is the typical biomass waste product of the furniture industry. Table 1 shows the proximate and ultimate analyses of pine sawdust. The pine sawdust was dried in air. The moisture content was approximately 10.4% in its air-dried basis. In a fixed bed reactor, first the moisture was removed and then the volatile. In its dry, ash-free basis, the carbon content of the sawdust was approximately 52.3%. The carbon in the sawdust reacted with oxygen in the air and steam to produce H₂, CO and CO₂. To produce a hydrogen-rich biogas, the gasification temperature was approximately 950 °C, the air equivalence ratio was 0.2 and the steam to biomass ratio was 0.8.

The composition of the biogas from the fixed bed reactor is shown in Table 2. The hydrogen content of the biomass gas reached 50.2% in volume, which is very beneficial for the operation of SOFCs. CO and CH₄ are also fuels for SOFCs, and CO₂ can participate in the catalytic reaction of an anode as the reforming gas [35]. Because the nitrogen is not involved in the reaction of SOFCs, we re-calculated the composition of the biogas without N₂, as shown in Table 2. The simulated biogas was prepared based on the composition of biomass gas without N₂. The

sulfur, nitrogen oxide and tar in the biogas were not measured and did not exist in the simulated biogas. To purify the biomass gas, we passed it through a serpentine tube immersed in ice water.

3.2. Biogas reforming

As the fuel of an SOFC, the biogas directly contacts the anode, where it oxidizes. For the biogas, the transformation of CH₄ and CO is the most pressing problem. When water is introduced (steam reforming), the reactions at the anode are very complex. The following reactions may occur simultaneously:



Due to these reactions, excess H₂ was produced. The main active substance of the anode material in a reduction state is the metallic nickel, which is an excellent catalyst for many reactions, especially at high temperatures. In order to examine the reaction process of biogas with the anode catalyst, we tested the catalytic performance of the anode material, Ni-YSZ, for the simulated biogas, especially the conversion process of methane. Fig. 2 shows the catalytic activities of the Ni-YSZ anode for the steam reforming of simulated biogas at different temperatures. It can be seen that the CH₄ conversion and CO selectivity greatly increased with the increase of temperature. Due to the sealing of

Table 1
Proximate and ultimate analyses of pine sawdust

Proximate analysis				Ultimate analysis					
M _{ad} /%	V _{ad} /%	A _{ad} /%	FC _{ad} /%	C _{daf} /%	H _{daf} /%	O _{daf} /%	N _{daf} /%	S _{daf} /%	
10.4	70.4	1.3	17.9	52.3	5.8	40.1	1.7	0.1	

Note: M: moisture, V: volatile, A: ash, FC: fixed carbon, ad: air-dry basis, daf: dry ash-free basis.

Table 2
The composition of the biogas

	H ₂	CO	CO ₂	CH ₄	N ₂
Biogas/%	50.2	10.4	11.0	9.8	18.6
Biogas (without N ₂)/%	61.7	12.8	13.5	12.0	/
Simulated biogas/%	61.0	13.0	14.0	12.0	/

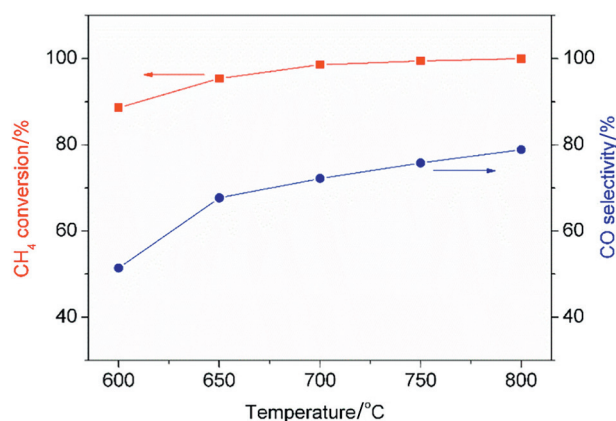


Fig. 2. Catalytic activities of the Ni-YSZ for the steam reforming of simulated biogas.

planar SOFCs, 750 °C is their common operating temperature, at which temperature the conversion ratio of CH₄ and the selectivity ratio of CO in biogas reached 99.5% and 75.8%, respectively. In the case of a fuel cell, the anode will produce a large amount of water. In this way, water will further participate in the catalytic conversion process of CH₄, which leads methane to have a higher conversion rate.

3.3. Biogas-fueled anode-supported SOFC

The anode-supported SOFC has a higher output than an electrolyte-supported one because of the lower resistance. The SOFC used in this work was prepared *via* tape casting and spraying technology. The microstructure of the anode was optimized. Fig. 3 shows the SEM images of cross-sections of an anode-supported SOFC after the electrochemical performance testing. Fig. 3a is an overview of cross-sections of the anode-supported SOFC. It can be seen that the electrolyte of the SOFC was very dense and thin. The anode and cathode were sintered on the opposite sides of the electrolyte, respectively. No obvious crack was observed between the electrode and electrolyte. The anode was very porous after the reaction. The cathode layer also presented a porous morphology. It seems that the contact between the current collector and the cathode is not very good. The SOFC was directly removed from the furnace after the cell performance test at the final operating temperature (600 °C), so an inconsistent contraction between these two layers happened due to the sudden cooling of the cell, resulting in a poor contact between the current collector and the cathode. The cooling rate will be strictly controlled in the practical application to avoid any peeling-off between different cell components.

The SDC barrier layer between the cathode and the electrolyte can be observed, and the thickness was only approximately 2 μm. This is very important for the high output performance of fuel cells. The magnified morphology of the anode in Fig. 3b clearly distinguishes between YSZ and Ni. The YSZ displays a continuous structure, while the Ni, distributed on the skeleton of YSZ, presents greater porosity.

To generate electricity, the fuel (hydrogen or biogas) was introduced into the anode of the SOFC and the cathode was exposed to the ambient air. To confirm the performance of the SOFC, H₂ was first used as the fuel. As shown in Fig. 4a, the cell generated OCVs of 1.081, 1.088, 1.092, 1.105 and 1.112 V at 800, 750, 700, 650 and 600 °C, respectively. The high OCVs proved that the electrolyte of the SOFC was dense and that the seal was good. When the SOFC started to generate electricity, the voltage of the cell decreased with the increase in current due to the existence of the internal resistance of the cell. At the appropriate voltage, the peak power density was obtained, which were 1753, 1596, 1328, 899 and 695 mW cm⁻² at 800, 750, 700, 650 and 600 °C, respectively. Fig. 4b shows the cell performance using the simulated biogas as the fuel. It should be noted that the simulated biogas was applied herein

because the quantity of biogas generated in our lab was too limited to feed SOFC for several hours. The peak power densities of 1702, 1521, 1328, 741 and 580 mW cm⁻² were achieved at 800, 750, 700, 650 and 600 °C, respectively. Similar high OCVs and power outputs were obtained when the SOFC operating on both fuels. It implies that the biogas derived from pine sawdust biomass will be a potential promising fuel for SOFCs. In order to verify this, the performance of SOFC operating on the generated biogas was tested, but at 750 °C solely due to its limited collected quantity. Fig. 5 shows the I-V polarization curves of the SOFC operating on various fuels at 750 °C. The peak power densities were 1596, 1391 and 1521 mW cm⁻², respectively, with hydrogen, biogas and simulated biogas as fuels. Because of the high content of hydrogen in the obtained biogas, the cell performance when operating on the biogas was close to that of simulated biogas, and slightly lower than that of hydrogen. The OCV of the cell with biogas as its fuel was 1.044 V, which was lower than both hydrogen and simulated biogas, and that may have been due to the dilution effect of nitrogen.

To obtain more information, the electrochemical impedances of the SOFC at 750 °C under OCV conditions were measured, with the results being shown in Fig. 6. Typically, there are two points of intersection with the Z' axis in the impedance curve. The left one (at high frequency) represents the ohmic resistance (electrolyte resistance). The right one (at low frequency) corresponds to the total resistance of the SOFC, which includes the ohmic resistance, the concentration polarization (mass-transfer or gas-diffusion polarization) resistance and the effective interfacial polarization resistances associated with the electrochemical reactions at both electrode-electrolyte interfaces

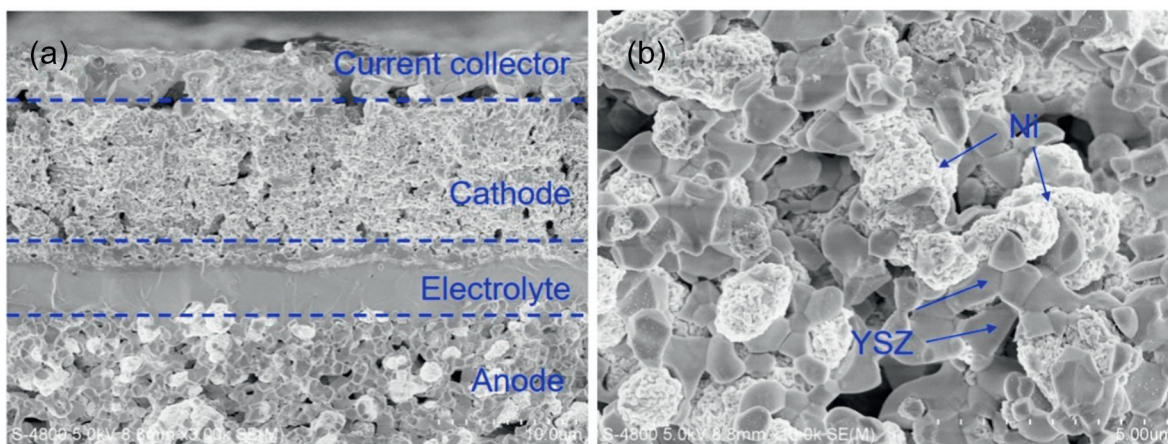


Fig. 3. SEM images of cross-sections of the SOFC: (a) overview and (b) anode.

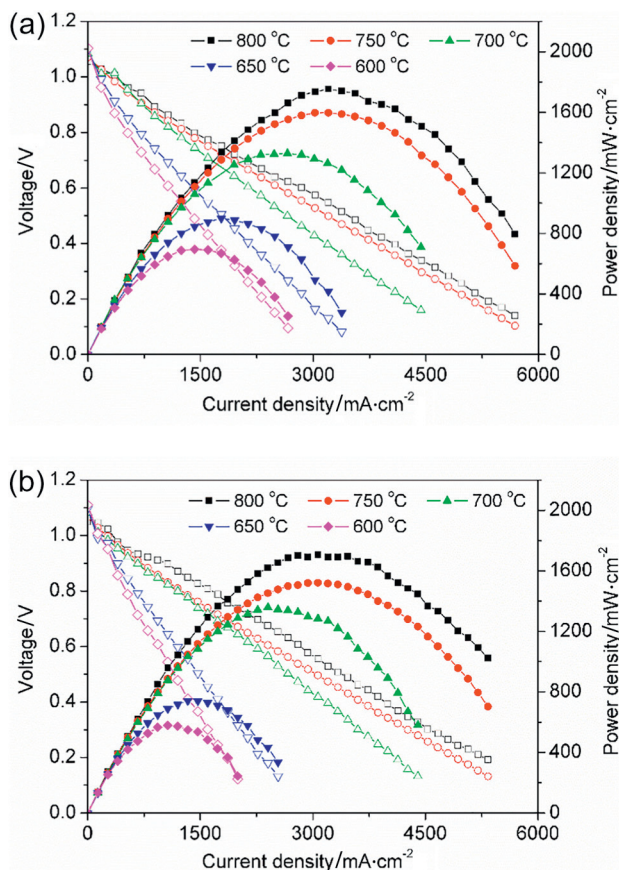


Fig. 4. I–V polarization curves of the SOFC operating on (a) hydrogen and (b) simulated biogas, respectively.

(anode-electrolyte and cathode-electrolyte) [42]. The ohmic resistances were 0.12, 0.16 and 0.19 $\Omega \cdot \text{cm}^{-2}$, respectively, operating on hydrogen, simulated biogas and biogas. Theoretically, the ohmic resistance should be the same due to the same configuration of SOFCs; however, the difference in values of resistance was found because the cells came from the different batches. Nonetheless, they are still similar. The corresponding electrode polarization resistances were 0.18, 0.18 and 0.23 $\Omega \cdot \text{cm}^{-2}$, respectively. Since the same configuration of SOFC was used in this study, the difference in electrode polarization resistance is mainly from the anode due to the various fuel components. The

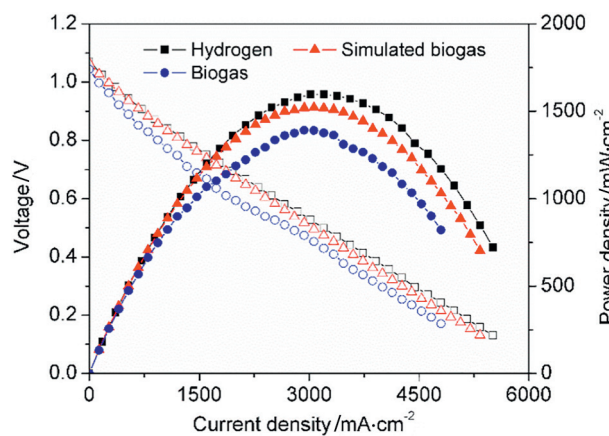


Fig. 5. I–V polarization curves of the SOFC operating on hydrogen, biogas and simulated biogas at 750 °C, respectively.

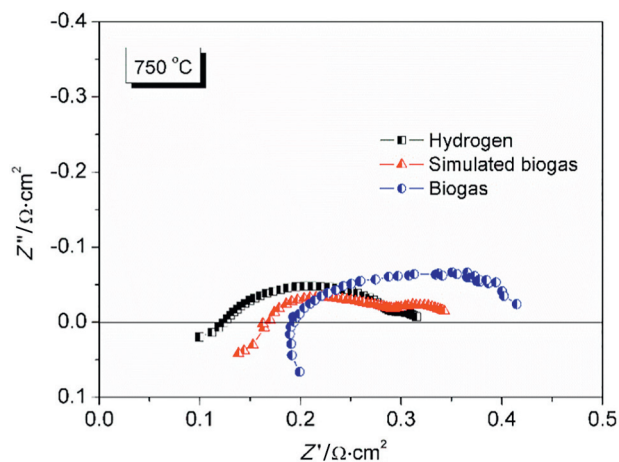


Fig. 6. Impedance spectra of the SOFC operating on hydrogen, biogas and simulated biogas at 750 °C, respectively.

polarization resistance slightly increased operating on biogas as compared to hydrogen and simulated biogas. The EIS results are in good agreement with I–V polarization results (Fig. 5). This indicated that the electrode polarization resistance played a dominant role in the electrochemical performance loss for the current anode-supported SOFC.

The stable operation of an SOFC is very important, especially when using a carbon-based fuel. If such fuel is cracked, the carbon that is produced will destroy the structure of the anode and thus decrease its mechanical strength. Fig. 7 shows the stability experiment results for the SOFC using hydrogen and simulated biogas as fuels, respectively. The current density of the stability test was 1800 $\text{mA} \cdot \text{cm}^{-2}$, and the operation time was 120 h. As shown in Fig. 7, the performance of the fuel cell was stable when using two kinds of fuel gas, indicating that the cell can withstand a short period of continuous work. After the 60 h operation, the performance losses of the SOFC were 0.8% and 1.2%, respectively, for hydrogen and biogas as fuels. Due to the limitations of laboratory conditions, the stability experiment was not carried out for a longer time. In the future, more attention should be paid to this for industrial applications.

4. Conclusions

In this study, the pine sawdust was gasified at 950 °C to obtain the biogas, which consisted of H_2 , CO, CO_2 and CH_4 . The content of H_2 was

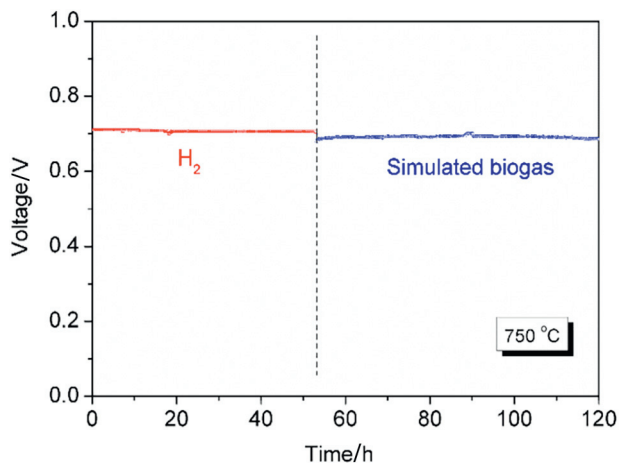


Fig. 7. Stability test of the SOFC fueled by hydrogen and simulated biogas at 750 °C.

50.2%. The results demonstrated that the biogas at the anode side continued reforming, and most of the CH_4 was converted to CO. High CO selectivity means greater H_2 generation. The maximum power density of the SOFC, with biogas and simulated biogas as the respective fuels, reached 1391 and 1521 mW cm^{-2} at 750 °C. The preliminary stability testing of the SOFC using simulated biogas showed that the biogas with high H_2 content could ensure the stable operation of the SOFC. The improvement in the integration of biomass gasification unit with SOFC system is needed to continuously generate electricity when biomass product gas is used as the fuel.

Acknowledgements

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