



Study on the preparation and production factors of a direct lignocellulose biomass fuel cell

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ABSTRACT

Lignocellulosic biomass is the most abundant renewable resource in nature. However, low-temperature fuel cell technology has not yet been able to use it as an efficient fuel. In this research study, a low temperature direct biomass fuel cell, which did not use enzymes or microorganisms as catalysts, was demonstrated using barley straw as the fuel, methyl violet as an electron mediator, nickel foam as an anode, and carbon cloth supported 0.15 mg/cm² of 10 wt% Pt/C doping graphene as a cathode. The effects of the particle sizes of the raw materials on the performance of the fuel cell were examined using 3% (w/v) different particle sizes of barley straw powder in 5 M NaOH and 0.02 M MV solutions. It was found that the smaller the particle size of barley straw powder was, the higher the electrochemical performance of the fuel cell would be. In addition, this study also examined the effects of several dissolution systems on the electrochemical performance of the fuel cell, including a 10 wt% LiCl/DMAC, 5 M NaOH, 7 wt% NaOH/12 wt% Urea, and 65 wt% ZnCl₂. It was found that the fuel cell in the 65 wt% ZnCl₂ solvent had the best electrochemical performance, and when the MV concentration increased, the electrochemical performance of the fuel cell also increased accordingly. The maximum power density of the fuel cell reached to 0.3 mW/cm² when the concentration of MV increased to 0.08 M, which was nearly 40 times higher than the microbial fuel cells fed with cellulose. Meanwhile, the open circuit voltage was 541 mV, and the limit current density was 0.3 mA/cm². This allowed the raw materials of the fuel cell to take a further step toward a more complex lignocellulosic biomass.

1. Introduction

Energy is a kind of important resource of living and developing for human. However, with the depletion of fossil energy and environmental crisis, the contradiction between supply and demand of energy is becoming more and more serious. In order to counter the growing threats of an energy crisis, as well as worsening environmental pollution problems, it has become necessary to access clean, affordable, and reliable energy sources. This has been the cornerstone of the social development and economic growth of the world since the Industrial Revolution. It has been known that biomass is the most abundant renewable resource, and it is included in a wide range of plants, such as crops, and waste. Biomass as a kind of clean energy has increasingly become one of the alternative energy sources for sustainable development [1–5]. Lignocellulose is a large species of biomass, and it includes the biomass which is composed of lignin, cellulose, and hemicellulose. It provides a plentiful and renewable resource for fuel and chemicals. It is mainly composed of woody, herbaceous plants which have a short growth cycle. However, it is difficult to obtain energy from lignocellulose at

low temperatures. A number of conventional techniques have been reported which have been able to utilize this energy, including saccharification, fermentation, and energy conversion. However, technical conditions and economic problems limit [6–11]. Also faced is the important problem of whether, after a variety of methods of pretreatment are completed, the energy of the biomass will be gradually lost according to the law of step-by-step decreasing energy.

Another method by which to use lignocellulose is to put it into fuel cells to obtain energy directly. Although the low-temperature fuel cell technology driven by enzymes or microbes has been rapidly developed, these cells have high purity requirements for raw materials, and can only selectively decompose specific biomass with low electric power output. Additionally, they have been found to lack stability and longevity, and require high maintenance for the bacterial cultures or immobilizations of the enzymes. Therefore, these current deficiencies hinder their applications [12–18]. Alternatively, direct biomass fuel cells have also been developed, the reaction mechanisms of which need to be supported by a large number of noble metals. However, relying on noble metals is not a long-term effective method according to the

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economic situation and low conversion rate. Related studies have shown that solid oxide fuel cells (SOFC) and direct carbon fuel cells (DCFC) use lignocellulosic biomass as fuel and can also reduce the dependence on noble metal catalysts [19–24]. However, they all require very high operating temperatures, as well as specialized equipment to maintain the energy to run entire systems and infrastructures. It has been found that, the SOFC are very susceptible to sulfur contaminants and carbon accumulation [24]. Also, the constructions of high-performance DCFC have some special requirements for the physical and chemical properties of the fuel, and due to this, further improvements in DCFC performances have been dependent on the regulation of the fuel characteristics [25,26]. Recently, it has been reported that electricity can be directly generated from raw biomass in solar-induced fuel cells catalyzed by polyoxometalate (POM) solutions. This kind of fuel cells with light or heating have obvious advantages such as energy saving and emission reduction compared to other forms. However, if there is no light irradiation or heating, the reduction of POM cannot occur, thus the power output is very low [27–29].

In this study, barley straw was placed in an air cathode low temperature fuel cell without using enzymes or microorganisms as a catalyst, instead of using a very small amount of 0.15 mg/cm² of 10 wt% Pt/C doping the oxide graphene as a catalyst supported with a carbon cloth as a cathode. We studied the power generation performances of barley straw fuel cells in different solvent systems and the effects of barley straw particle size and concentration of the MV on the electrochemical performance. We attempted to tap the power generation potential of lignocellulose biomass.

2. Experimental

2.1. Materials

In this study, barley straw was used as the fuel, which was collected from China's Guizhou Province. Methyl viologen (MV, Yixiubogu Biological Technology Co., Ltd., Beijing, China) was used as the electron mediator. Carbon cloth (HCP330) was used as a substrate for the catalyst as a cathode. Nickel foam (PPI-70-110, Han Bo Environmental Protection Equipment Co., Ltd., Shenzhen, China) was used as an anode. Carbon black powder (Cabot Vulcan XC-72), along with a 60% weight PTFE solution (Aldrich PTFE 60 wt% dispersion in water), was used to construct the cathode support layer. The 10 wt% Pt/C, 5 wt% Nafion solution, pure isopropanol (Hesen Technology Co., Ltd., Shanghai, China) and graphene were necessary materials for making the air cathode. All of the solutions were prepared using ultrapure water.

2.2. Preparation of the biomass fuel cells

As previously described, a single chamber direct lignocellulosic fuel cell was employed [30–32]. It was constructed from polymethylmethacrylate (PMMA). The air-breathing carbon cloth cathode and nickel foam anode were placed on both sides of a 30 mm diameter cylindrical internal chamber with a volume of 12 mL. The nickel wire penetrated both sides of the middle plate, and spanned across the chamber diameter position to contact the electrodes closely. The plate of the cathode side was not sealed, thus exposing the oxygen to act in the reaction. The design process of the three-layer air-breathing O₂-reduction cathode was referred to the previous method [33]. The difference was that we reduced the amount of catalyst 10% Pt/C from the conventional of 0.5 mg/cm² to 0.15 mg/cm² by doping RGO (reduced graphene oxide, 0.1 mg/cm²). The RGO was prepared by using hydrazine hydrate to reduce the graphene oxide, and the graphene oxide was prepared by the Hummers method [34]. RGO had a two-dimensional structure with a large theoretical surface area, strong electrical conductivity, and good corrosion resistance. These excellent performances allowed the graphene can be used as a promising catalyst

carrier in the fuel cells [35–38]. In the future, there may be a co-catalysis between the surface active center of the RGO and the Pt catalyst, which may make the RGO become the main catalyst carrier [39–41].

2.3. Treatment of barley straw

A high-speed universal crusher (FW100, Tianjin Tai Site Instrument Co., Ltd.) was used to crush barley straw into powders of different particle sizes; the crusher included the mesh sizes of more than 40 mesh, 40–60 mesh, 60–80 mesh, 80–100 mesh, and less than 100 mesh. Then the 80–100 mesh size barley straw powder was dissolved using four different solvent systems. One was the ionic dissolution system; some studies have shown that 65 wt% concentration of ZnCl₂ solution dissolves cellulose best. At this concentration, the binding of zinc ions to water is weakened and becomes more strongly bound to the hydroxyl groups on the cellulose, thereby resulting in the dissolution of cellulose. In the same way, the other three dissolution systems were selected to determine which system of barley straw fuel cell power generation performance was the best. Studies have shown that 7 wt% NaOH/12 wt% urea, 5 M NaOH, 10 wt% LiCl/DMAc also have a good solvent effects on cellulose. First, barley straw were suspended in 5 M NaOH solution and 7 wt% NaOH/12 wt% urea solution for shaking and ultrasound at room temperature, respectively. Both were stored at –20 °C until they were equilibrated at this temperature. Second, a barley straw sample stirred in 10 wt% LiCl/DMAc solution and 65 wt% ZnCl₂ solution for 120 min respectively, then it allowed to stand still for 24 h at room temperature. The barley straw concentration in all of the above solutions was 3% (w/v).

2.4. Electrochemical characterization and fuel cell tests

In this study's experiment, the NI6009-USB (Shenzhen, China) acquisition system was used to measure and record the voltage of the fuel cell. When the output voltage of the cell ran to a stable stage, the external circuit was disconnected and the cell was left for 0.5 h before the OCV was recorded. The polarization and power density curves were measured using electrochemical workstations, or by changing the external resistance method. However, some studies have shown that the scanning rate of electrochemical workstations will affect the power output [42]. Therefore, we used the steady-state discharge method to determine the cell power conditions. After measuring the open circuit voltage, we connected a sliding rheostat to both ends of the cell and changed the external resistance variations from 10 to 9999 Ω. The cells were left to stabilize at each resistance for 5 min before the voltage measurement was taken. Then, the power density (W/m²) was obtained according to cell voltage (V), current (I) which can be calculated by Ohm's law. The cyclic voltammogram (CV) was carried out utilizing an electrochemical workstation (CHI660D, Chenhua Technology Co. Ltd., Shanghai, China). A saturated calomel electrode was used as the reference electrode, and a platinum wire was used as the counter electrode. The samples were scanned at 50 mV/s ranging from –1.4 to 0.8 V. The test temperature of the CV and cell performance was 25 °C.

2.5. Surface structure analysis using scanning electron microscopy (SEM)

After a gold spraying process, the images of the surface structure changes of barley straw particles before and after the reaction were obtained using an S-3000N SEM device (Hitachi, Tokyo, Japan).

2.6. Product identification

The discharge curve of the fuel cell connected to a resistance of 1000 Ω as the load was measured over time until the voltage was no longer detectable. Then the liquid part of the reaction remaining material was measured by a liquid-state C13 NMR (JNM-ECA600, JEOL, Japan) and the solid residues ground to a powder with a certain amount

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