



Electrochemical gas–electricity cogeneration through direct carbon solid oxide fuel cells



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H I G H L I G H T S

- Electricity and CO can be cogenerated through a direct carbon solid oxide fuel cell.
- Operating time can be used to characterize the rate decrease of Boudouard reaction.
- CO production decreases with operating time while CO₂ increases.
- Overall efficiency is twice of electrical one when CO is taken as useful output.
- Electrical efficiency increases with run time while overall efficiency decreases.

A R T I C L E I N F O

Article history:

Received 21 August 2014

Received in revised form

23 November 2014

Accepted 4 December 2014

Available online 4 December 2014

Keywords:

Gas–electricity cogeneration

Direct carbon

Solid oxide fuel cell

Carbon monoxide production

Boudouard reaction

A B S T R A C T

Solid oxide fuel cells (SOFCs), with yttrium stabilized zirconia (YSZ) as electrolyte, composite of strontium-doped lanthanum manganate (LSM) and YSZ as cathode, and cermet of silver and gadolinium-doped ceria (GDC) as anode, are prepared and tested with 5wt% Fe-loaded activated carbon as fuel and ambient air as oxidant. It is found that electricity and CO gas can be cogenerated in the direct carbon SOFCs through the electrochemical oxidation of CO and the Boudouard reaction. The gas–electricity cogeneration performances are investigated by taking the operating time of the DC-SOFCs as a measure of rate decrease of the Boudouard reaction. Three single cells and a two-cell-stack are tested and characterized in terms of electrical power output, CO production rate, electrical conversion efficiency, and overall conversion efficiency. It turns out that a rapid rate of the Boudouard reaction is necessary for getting high electrical power and CO production. Taking the emitted CO as part of the power output, an overall efficiency of 76.5% for the single cell, and of 72.5% for the stack, is obtained.

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1. Introduction

Carbon, as the main component of fossil fuel (coal, oil, and natural gas) and biomass, is an important energy carrier. Conventionally, the energy stored in carbon-based fuels is released through combustion to get heat which is then converted to mechanical or electrical energy for terminal applications. Such combustion involves mixing fuel and air at very high temperature and

hazardous products, such as NO_x, are formed and emitted, resulting in air pollution [1–4]. Meanwhile, the energy conversion efficiency is limited, leading to significant waste of fuel and large production of green house gases which cause the global climate change, not to mention the depletion crisis of fossil fuel reserves [5]. Thus, alternative conversion ways are considered to increase the efficiency and reduce the pollution in converting the chemical energy of carbon-based fuels to the energy for terminal application, generally electricity. Electrochemical conversion might be a promising option.

A device, consisting of electrolyte, cathode, and anode, that can directly convert the chemical energy stored in fuels to electrical

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power, is called a fuel cell [6,7]. Fuel cells operated on hydrogen fuel have been studied widespread. Carbon-based fuels, such as hydrocarbons, carbon monoxide, and solid carbon have been applied as fuels of high temperature fuel cells with molten carbonates/hydride or solid oxide as electrolytes [8–10]. Direct carbon fuel cells (DCFCs), within which, carbon is the fuel feedstock, have been attracting more attention due to their high conversion efficiency and fuel utilization as well as abundant carbon sources, such as coal and biomass [8–10].

Theoretically, the chemical–electrical conversion efficiency of carbon through the complete oxidation reaction, $C + O_2 = CO_2$, is the Gibbs energy difference of the reaction divided by the enthalpy, $\Delta G/\Delta H$. The absolute value of ΔG is a little bit larger than that of ΔH , resulting in the efficiency higher than 100%, meaning that some external heat provided to the reaction may be converted to electricity. For example, the absolute values of ΔG and ΔH at 800 °C are 395.56 and 394.37 kJ mol⁻¹, respectively, thus the theoretical efficiency is 100.3%. Meanwhile, the fuel utilization of DCFC is high because carbon is in solid state and its oxidation products (CO and CO₂) are in gas phase so that the reaction is not limited by thermodynamic equilibrium and carbon can be completely used up in the chemical–electrical conversion.

A solid oxide fuel cell (SOFC) [11], with an oxygen ion conducting oxide membrane as electrolyte, is a whole solid state device that can be operated with solid carbon as fuel. Such a SOFC with carbon used as fuel feedstock is generally called as direct carbon SOFC (DC-SOFC). Here, the word “direct” means direct utilization rather than direct oxidation because carbon is not directly oxidized in the cell, as will be explained later. A DC-SOFC may give an output comparable with that operated on hydrogen [12–18]. Fig. 1 shows a schematic illustration of a DC-SOFC setup. Carbon is not necessary to have contacts with the anode. Actually, it is delivered through the reversed Boudouard reaction which produces CO for the anode reaction. In other words, CO is electrochemically oxidized at the anode, by oxygen ions coming through the electrolyte. The DC-SOFC provides an efficient way to convert the chemical energy of carbon into electricity [10,18–22]. In fact, it is a membrane reactor in which the electrolyte membrane only permits oxygen ions to go through and the emitted gases from anode are only CO and CO₂. Nitrogen, which occupies large fraction volume of the emission of combustion, does not appear in the anode emission of a DC-SOFC because it is filtered out by the electrolyte membrane. Without N₂ involved, CO and CO₂ can be separated and handled with much lower cost [23,24].

While in principle solid carbon can be completely used up in its oxidation, it is generally partly oxidized because CO is the favored product at high temperature rather than CO₂ [10,18]. This means that in the electrochemical utilization of carbon, the

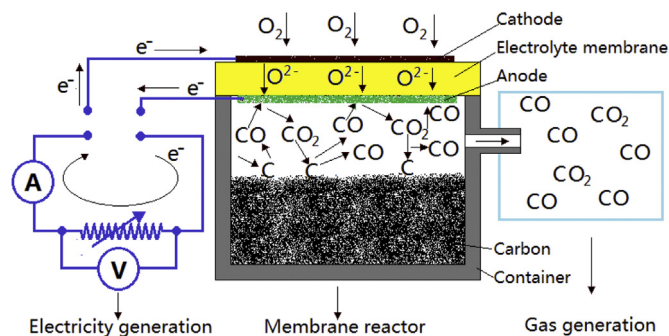


Fig. 1. Schematic illustration of electricity–gas cogeneration through a direct carbon solid oxide fuel cell.

chemical–electrical conversion efficiency is much lower than the theoretical $\Delta G/\Delta H$. Nevertheless, pure CO is a valuable product which can be used as high quality fuel [25], chemical feed stock, and especially as the necessary component of syngas used in industrial Fischer–Tropsch synthesis [26], etc. Therefore, the DC-SOFC shown in Fig. 1 can be taken as an electricity–gas cogeneration system.

In this paper, we present our work on electricity–gas cogeneration through DC-SOFCs. The performances of the DC-SOFC systems are investigated through characterizing the electrical power output, CO production rate, electrical conversion efficiency, and overall conversion efficiency. Correlations between the operating parameters are analyzed in details, according to theories of thermodynamics and kinetics.

2. Experimental

2.1. Fabrication of tubular electrolyte membranes

Tubular electrolytes of 1 wt% Al₂O₃-doped-YSZ were fabricated by dip coating technique [27–29]. 25 g YSZ powder (TZ-8Y, Tosoh Corporation, Tokyo, Japan) was mixed with 0.25 g Al₂O₃ (1 wt%) (Xinfumeng, China, with a first modal particle size of 0.243 μm (12.67 vol.%) and second 1.94 μm (8.25 vol.%)). A homogeneous electrolyte slurry was prepared by ball milling the mixed powder for 3 h, with 0.8 g dioctyl phthalate (DOP) and 0.8 g polyethylene glycol (PEG) as plasticizing agent, 0.8 g triethanolamine (TEA) as dispersant, 1.8 g polyvinyl butyral (PVB) as binder, and 42 g ethanol as solvent. Then the slurry was poured into a beaker. A glass test tube, used as a mould, was dipped into the slurry vertically. Two seconds later, the mould with a layer of the slurry coated on it, was taken out and dried in air. After the ethanol in the layer was volatilized, a green electrolyte layer was formed on the mould. This dip-coating process was repeated for 10 times to get a required thickness of the coated layer. Then the layer was removed from the mould and sintered at 1400 °C in air for 5 h to form a dense electrolyte membrane tube [30]. The sintered tubes were 1.10 cm in diameter, 1.50 cm in length and 0.16 mm in thickness. Some tubes with two ends open were prepared in a similar way, only the closed end of the coated layer had been cut off before it was removed from the mould and sintered.

2.2. Cell assembling

A composite cathode paste composed of 1.2 g LSM (La_{0.7}Sr_{0.3}MnO₃, Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences) and 0.8 g YSZ (Tosoh) was prepared by mixing the LSM and YSZ powders in an agate mortar and grinding the mixture for 3 h with 3 g PVB terpeneol solution of 10 wt% concentration as binder [31]. The same process was used for preparing a pure LSM paste. A composite LSM-YSZ functional layer and a pure LSM current collecting layer were sequentially applied on the outside of the wall of each electrolyte tube by brush painting with the pastes, and then were dried and co-sintered at 1100 °C for 2 h.

A cermet of Ag and GDC (gadolinium doped ceria, Ce_{0.8}Gd_{0.2}O_{1.9}) was used as the anode material because Ag is a good electronic conductor and has high catalysis activity to promote the oxidation of CO, while GDC, with high ionic conductivity, combined with Ag, can provide more three phase boundaries for the electrochemical oxidation of CO [19–22]. 1.75 g sliver paste (DAD-87, Shanghai Research Institute of Synthetic Resins, Shanghai, China, containing 80 wt% Ag), 0.6 g GDC powder (Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences), and 3 g 10 wt% PVB terpeneol solution were mixed by grinding the mixture

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