Journal of Power Sources 371 (2017) 106–111

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Durability and robustness of tubular molten carbonate fuel cells

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HIGHLIGHTS

- Tubular molten carbonate fuel cells were built and tested.
- Tubular MCFC is more durable and robust than planar MCFC.
- Tubular MCFC can tolerate a differential pressure between anode and cathode 0.1 MPa.
- Tubular MCFC can tolerate a rapid temperature change of about 100 °C per hour.
- Tubular MCFC can tolerate H_2S levels in the fuel gas of up to 20 ppm.

ARTICLE INFO

Keywords:
Molten carbonate fuel cell
Tubular type
Li/Na carbonate electrolyte
Pressurization system
Hydrogen sulfide

ABSTRACT

One anticipated system for high-efficiency power generation is the combination of syngas from gasification and high temperature fuel cells. The system uses a pressurization system, and it takes into account poisoning by impurities in the syngas. The durability and robustness of the fuel cells used in this system are an important issue for commercial applications. This study focuses on tubular molten carbonate fuel cells (MCFCs), which seem to be relatively durable compared with conventional planar-type MCFCs. Various power generation tests were performed in order to evaluate the durability and robustness of the tubular MCFCs. After continuous generation tests at 0.3 MPa, the cell voltage decay rate was found to be 0.8 mV/1000 h at 0.2 A/cm^2. Moreover, it was found to be possible to generate power stably with fuel gas containing 20 ppm H_2S. When the differential pressure between the anode and cathode was set to 0.1 MPa, the power generation tests were performed without gas leakage. In addition, starting (heating) and stopping (cooling) could be done in a short period, meaning that the cold start/stop characteristics are favorable. Therefore, the tubular MCFC was confirmed to have the durability necessary for a power generation system.

1. Introduction

Gasification fuel cell generation systems that gasify resources such as coal and biomass as fuel are expected to become one type of system used for high-efficiency power generation in the future [1–4]. Examples of the fuel cells include molten carbonate fuel cells (MCFCs) and solid oxide fuel cells (SOFCs), which can use hydrogen and carbon monoxide as fuel, although problems such as durability of the fuel cells and production costs of the systems remain for practical implementation of these systems. Furthermore, because integrated gasification fuel cell (IGFC) systems are more complicated than power generation using natural gas, the fuel cells need to be robust to various system faults. Additionally, the gases produced by the gasification of coal and biomass contain large amounts of impurity gases, namely, hydrogen sulfide and hydrogen chloride. It is therefore necessary to purify of the gases to within the tolerable limits of the fuel cells, with the purification cost depending on the level of gas purification. The durability of fuel cells against impurities thus requires close attention. This paper focuses on MCFCs, which are relatively durable against impurity gases. MCFCs can be divided into two types: internal-reforming MCFCs, which use methane as their primary fuel, and external-reforming MCFCs, which use syngas (H_2 and CO) as their primary fuel. Current implementations use internal-reforming MCFCs, as external-reforming MCFC suitable for gasification have not been implemented due to the high production costs and high total system costs of MCFCs.

We have successfully developed a tubular MCFC with low production costs that can overcome the above problems [5]. These tubular MCFCs do not have a separator and do not require high-precision manufacturing of the cell components. Furthermore, since there is no need to stack the cells and the cells are not readily affected by adjacent cells, it is not necessary to strictly account for thermal expansion due to temperature changes, thus giving the cells a wide range of operational control. These advantages of tubular MCFCs are expected to allow the construction of coal gasification MCFC systems with excellent

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http://dx.doi.org/10.1016/j.jpowsour.2017.10.024
Received 16 August 2017; Received in revised form 17 September 2017; Accepted 9 October 2017
Available online 21 October 2017
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Table 1
Content of power generation tests conducted on tubular MCFCs.

Cell No.	Electrolyte	Tests
1	Li_2CO_3/K_2CO_3 = 64/36	I–V characteristics, temperature dependence
2	Li_2CO_3/Na_2CO_3 = 53/47	Pressure dependence
3	Li_2CO_3/Na_2CO_3 = 53/47	Continuous generation test at 0.3 MPa, effects of differential pressure on the anode and cathode
4	Li_2CO_3/Na_2CO_3 = 53/47	I–V characteristics, temperature dependence
5	Li_2CO_3/Na_2CO_3 = 53/47	Cold start/stop characteristics
6	Li_2CO_3/Na_2CO_3 = 53/47	Evaluation of effects of H_2S in fuel gas on cell voltage

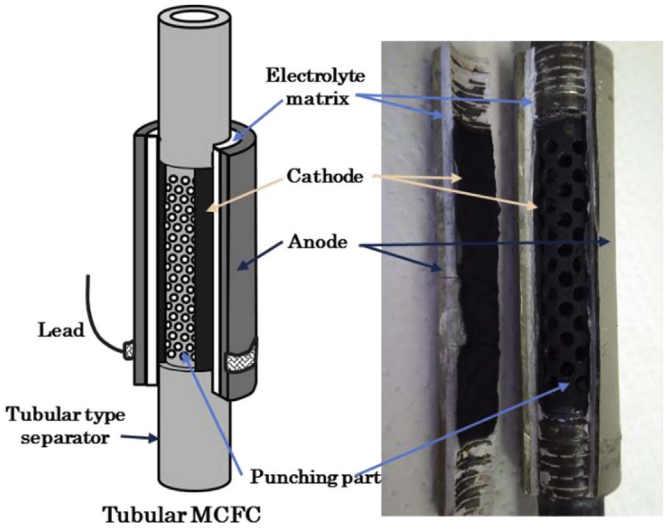

Fig. 1. Illustration of tubular MCFC.

operability. In this study, we evaluate the basic power generation performance and durability of tubular MCFCs in order to investigate their suitability for MCFC gasification systems.

2. Experiment

2.1. Preparation of tubular MCFCs

Cell performance data were obtained by using the six cells listed in Table 1. Fig. 1 shows a schematic illustration and a photograph of a cell. The manufacturing process for the tubular MCFCs was the same as the process described in our previous work [5]. The cathodes were made of lithiated NiO-3%MgO. A matrix made of $LiAlO_2$ supported the electrolyte. The anodes were made of Ni-2%AlCr. Two types of carbonate mixtures (Li_2CO_3/Na_2CO_3 = 53/47 mol % with a melting point of 496 °C and Li_2CO_3/K_2CO_3 = 62/38 mol % with a melting point of 488 °C) were used as the electrolyte. It was previously reported that the performance of cells using Li_2CO_3/Na_2CO_3 is higher than that of cells using Li_2CO_3/K_2CO_3 [6]. Therefore, Li_2CO_3/Na_2CO_3 was used as the main electrolyte.

The quantity of the electrolyte was set such that the electrolyte packing ratios (the volume of pores filled with electrolyte relative to the total pore volume) of the anode, cathode, and electrolyte matrix were 30–50%, 30–50%, and 100%, respectively.

When calculating the current density and other parameters, the electrode surface was assumed to be the contact surface between the cathode and the electrolyte matrix.

2.2. Power generation test apparatus

Fig. 2 shows the schematic illustrations of power generation test apparatuses A and B. Apparatus A was used to obtain data at atmospheric pressure. The internal dimensions of the electric furnace were an internal diameter of 140 mm and a height of 500 mm. The temperature of the electric furnace was controlled such that the temperature near the cell was 650 °C. The flow rate of a fuel gas mixture containing H_2, CO_2, and N_2 was regulated by a mass controller, and a bubbler was used to add water vapor. The flow rate of an oxidant gas composed of air and CO_2 was regulated by a mass controller and supplied to the cell. When the effects of H_2S on the cell performance were evaluated, gas cylinders containing N_2 and H_2S (500 ppm) were prepared and the flow rates of the main gas ($H_2/CO_2/H_2O$) and impurity gas (N_2/H_2S) were regulated for the target concentration of H_2S. The concentration of H_2S in the inlet and outlet gases was analyzed using a gas detector tube.

Apparatus B was used to obtain data under pressure. The internal dimensions of the electric furnace were an internal diameter of 70 mm and a height of 600 mm. The temperature control and gas flow control methods were similar to those used with apparatus A. The pressures of the fuel gas and the oxidant gas were controlled by pressure adjustment valves.

The gas composition inside the electric furnaces changes as fuel is consumed during power generation. However, the interior volumes of both electric furnaces are extremely large compared with the size of the cell, and the flow rate of the supplied fuel gas is small, so the gas inside the electric furnaces reaches an almost uniform condition at the steady state.

3. Results and discussion

3.1. Basic power generation performance

Power generation tests were performed with the tubular MCFC using Li/Na carbonate in order to obtain basic data (I–V characteristics, temperature dependence, pressure dependence, and long-term performance). Fig. 3 shows the basic performance of tubular MCFC.

3.1.1. I–V characteristics

Fig. 3(a) shows the changes in the current–voltage characteristics (I–V characteristics) of a Li/Na carbonate cell during power generation, with the I–V characteristics of a Li/K carbonate cell shown for reference. In order to collect these data, a current density of 200 mA/cm^2 was used as the standard to alter the current density quickly. Because a change in current density corresponds to an extremely small change in the amount of gas relative to the volume of the electric furnace, the fuel gas composition at all current densities is assumed to be equivalent to that at 200 mA/cm^2. As mentioned in previous works about planar MCFCs, the I–V characteristics of a Li/Na carbonate cell exhibit the same tendencies as those of a Li/K carbonate cell, and the performance of a Li/Na carbonate cell is higher than that of a Li/K carbonate cell.

3.1.2. Temperature dependence of cell voltages

Fig. 3(b) shows the effects of temperature on the voltages of a Li/Na carbonate cell and a Li/K carbonate cell. Because the cell inlet temperature is set at 580 °C and the cell outlet temperature at 660 °C in conventional planar MCFC stacks, the temperature characteristics for this test were acquired in the range 580–680 °C. As can be seen from the plot, the cell voltage of a Li/Na carbonate cell is higher than that of a

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