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Study and performance test of 10 kW molten carbonate fuel cell power generation system

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Abstract The use of high-temperature fuel cells as a power technology can improve the efficiency of electricity generation and achieve near-zero emissions of carbon dioxide. This work explores the performance of a 10 kW high-temperature molten carbonate fuel cell. The key materials of a single cell were characterized and analyzed using X-ray diffraction and scanning electron microscopy. The results show that the pore size of the key electrode material is 6.5 μm and the matrix material is α-$LiAlO_2$. Experimentally, the open circuit voltage of the single cell was found to be 1.23 V. The current density was greater than 100 mA/cm^2 at an operating voltage of 0.7 V. The 10 kW fuel cell stack comprised 80 single fuel cells with a total area of 2000 cm^2 and achieved an open circuit voltage of greater than 85 V. The fuel cell stack power and current density could reach 11.7 kW and 104.5 mA/cm^2 at an operating voltage of 56 V. The influence and long-term stable operation of the stack were also analyzed and discussed. The successful operation of a 10 kW high-temperature fuel cell promotes the large-scale use of fuel cells and provides a research basis for future investigations of fuel cell capacity enhancement and distributed generation in China.

Keywords Fuel cell stack · Key materials · Molten carbonate fuel cell · Power generation test

1 Introduction

In recent years, with restrictions of carbon emissions globally, increasing numbers of countries have changed their coal utilization technologies. China currently mainly uses coal resources and has a certain degree of dependence on oil and gas. It is particularly critical to improve the efficiency of coal power generation and reduce emissions of carbon dioxide and pollutants in the process of energy transformation (Mehrpooya et al. 2017; Zhang 2018; Xu et al. 2019). Fuel cell power generation technology (McPhail et al. 2015; Mastropasqua et al. 2019) has become established in the public consciousness and has been continuously developed by various countries across the world. Fuel cell power stations have been demonstrated in Europe, America, and elsewhere (Carapellucci et al. 2019). However, there is no stationary high-temperature fuel cell power station in China as yet, owing to a technological gap with other countries. Many scholars have begun research on various fuel cell technologies, such as proton exchange membrane fuel cells, solid oxide fuel cells (SOFCs), and molten carbonate fuel cells (MCFCs).

The technology of integrated gasification combined cycle (IGCC) power generation, based on integrated gasification fuel cells (IGFCs), can greatly improve coal power efficiency and carbon dioxide capture (Duan et al. 2015; Torabi et al. 2016; Dong et al. 2019; Wang et al. 2020) and achieve near-zero emissions of carbon dioxide and pollutants. IGCC power generation is still at a conceptual stage in China (Ku et al. 2018) but it may be possible to move

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toward a practical demonstration of IGFCs in the next decade, with current progress in fuel cell technology. IGFC systems based on MCFCs and SOFCs have been demonstrated (Campanari et al. 2016; Samanta and Ghosh 2017; Wolfersdorf and Meyer 2017; Mu et al. 2018; Slater et al. 2018), it is clear that the fuel cell integrated CO_2 capture process is a promising route, and will be more effective if fuel cell technology can be commercialized (Wang et al. 2020). The fuel cell, as a key power generation device, has the advantages of being very efficient and environmentally friendly. It can convert chemical energy to electrical energy directly, and can be used for refrigeration and heating by system coupling (Wu et al. 2016; Ghorbani et al. 2019; Marefati and Mehrpooya (2019). A theoretical energy conversion efficiency greater than 85% can be achieved, without the emission of nitrogen oxides or sulfur oxides. As a high-temperature fuel cell, MCFC has a wide range of fuel sources and does not rely on the use of precious metals as electrode catalysts. Moreover, MCFCs can be combined with gas or steam turbines to achieve combined heat and power, with improved energy utilization and conversion effectiveness (Tano and Makino 2017).

MCFCs are mainly used in large-scale power generation, distributed power generation, and fixed power systems. Among the various different types of fuel cell, they remain the largest single-unit installed capacity (Samanta and Ghosh 2016; Mastropasqua et al. 2017; Makino and Noda 2018). Moreover, they are held to be of great importance and have been developed in the USA, Germany, Italy, South Korea, Japan, and other countries (Mehmeti et al. 2018; Haghghi et al. 2019; More et al. 2020). Fuel cell Energy, in the USA, continues to research and develop fuel cell–gas turbine power generation systems with natural gas as a fuel, and has developed three commercial power plants utilizing MCFCs of 300 kW to 2.8 MW, namely, DFC300MA, DFC1500, and DFC3000. The maximum effective area of a single cell may be as much as 1 m^2; the current density of the stack is 80–120 mA/cm^2. The power generation efficiency is greater than 47%. In 1995, a 1 MW power station composed of four 250 kW fuel cell modules was built in Japan (Discepoli et al. 2016; Da Silva and Matelli 2019). This power station integrates coal gasification combined cycle power generation and coal gasification fuel cell combined cycle power generation (IGFCs), with the goal of achieving 55% power efficiency in the future. An MCFC power station (58.8 MW, comprising 21 fuel cell modules, each of 2.8 MW) was established by South Korea's POSCO in Gyeonggi Province, and can provide power for 140,000 households.

In 2017, China's Ministry of Science and Technology set up a major special project "CO_2 near-zero emission coal gasification power generation technology" to conduct research and demonstrations of MW-level thermal power IGFC power generation systems. The core of the project is to develop the first domestic 500 kW high-temperature fuel cell power generator. The goal of this research project is to develop domestic high-temperature fuel cell technology, accelerate the demonstration application of fuel cell power generation technology, and reduce the technological gap with other countries. The 500 kW high-temperature fuel cell consists of SOFCs and MCFCs. The 10 kW fuel cell described in this study will be used as the module unit for the MCFC.

In this study, a 10 kW MCFC stack, which is the leading domestic fuel cell technology, was designed and built. We studied the operation and performance of the fuel cell stack. Problems arising during operation of the stack were analyzed, and the results are discussed in terms of operation power and time. The operation of a 10 kW MCFC will increase the power and scale generation of domestic fuel cells and provide a research basis for fuel cell capacity enhancement in future work.

2 Experiments

2.1 Fuel cell materials

Molten carbonate fuel cells are mainly composed of electrodes, separators, metal bipolar plates, and electrolytes, as shown in Fig. 1a. The flow channel of the fuel cell in the stack is illustrated in Fig. 1b.

The anode material was porous metal nickel (Antolini 2011; Kulkarni and Giddey 2012) and the cathode was porous nickel oxide (generally obtained by oxidizing pure nickel during fuel cell heat treatment). The electrode was prepared using carbonyl nickel powder as raw material, adding sodium carboxymethyl cellulose solution as a binder. The slurry was cast on a flat surface by mixing and stirring, and the electrode blank was dried thoroughly. The electrode blank was placed in a high-temperature furnace for heat treatment. During the sintering process, the size and distribution of the pore size were controlled by controlling the temperature and pore-forming agent.

The matrix material in the MCFCs was α-$LiAlO_2$ (Zhang et al. 2017; Kim et al. 2019). An α-$LiAlO_2$ powder was prepared using a high-temperature roasting method. The process was as follows: Li_2CO_3 and basic alumina were mixed in an equimolar ratio. To complete the reaction, an excessive amount of Li_2CO_3 (2 wt%) was added. The matrix was then prepared as follows: solvent (*n*-butanol), binder (polyvinyl butyral), dispersant (fish oil), plasticizer (dioctyl phthalate), defoamer (silicone oil), and $LiAlO_2$ powder were ball milled according to the desired proportion, using distilled water as the ball milling

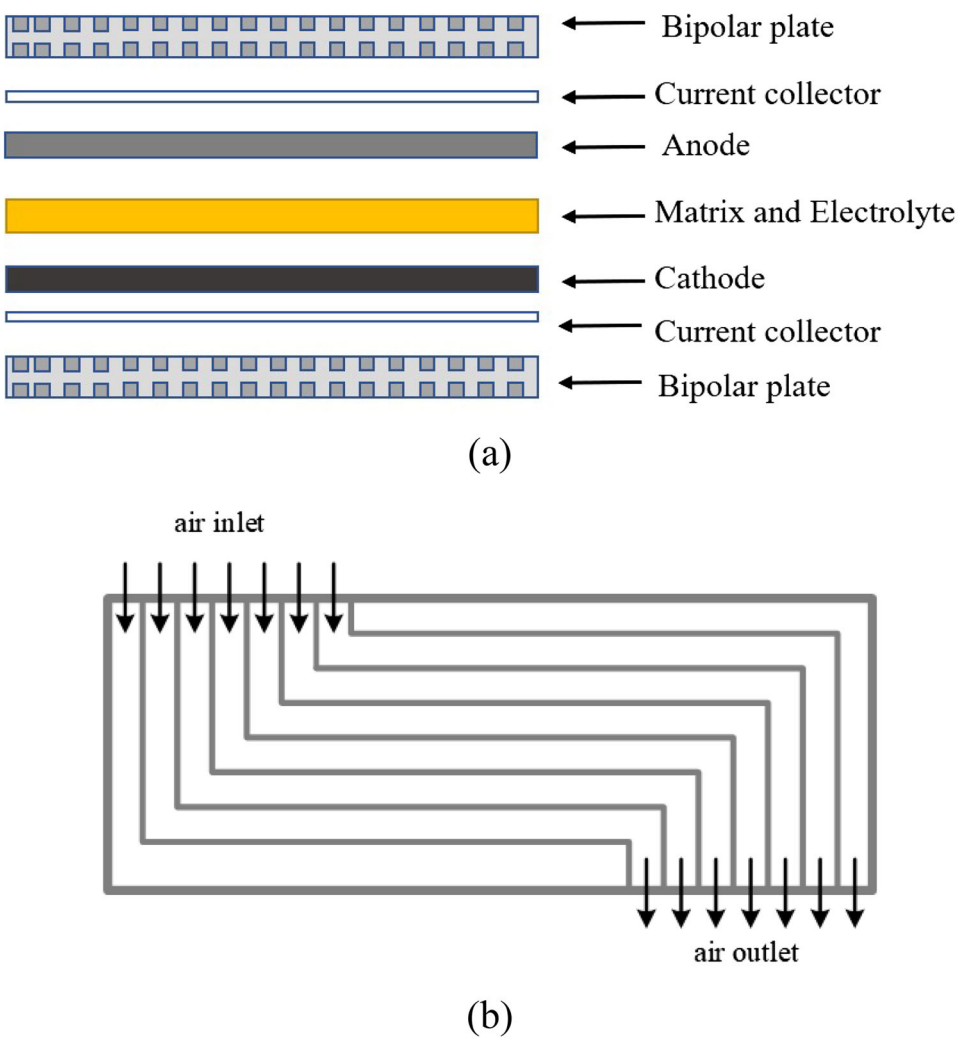

Fig. 1 **a** Structure of molten carbonate fuel cell; **b** flow channel

medium, and the ball mill rotated to form a uniform slurry. The obtained slurry was then subjected to vacuum defoaming, and a film casting method was used to form a film. After ball milling, drying, and roasting, α-$LiAlO_2$ was obtained.

The bipolar plate material was 316L stainless steel, and the processing method used a combination of stamping and welding. The electrolyte was Li_2CO_3/K_2CO_3 (62 mol%/38 mol%).

2.2 Fuel cell stack

The fuel cell stack and the single cell were assembled manually. Each cell had an anode and a cathode, and there was one bipolar plate between each cell, as illustrated in Fig. 1a. Each single cell was separated by a matrix. The 10 kW MCFC stack is shown in Fig. 2; Table 1 gives the fuel cell parameter data.

2.3 MCFC power generation system test

The test system for the MCFC stack is shown in Fig. 3. During the test, the fuel gases used were pure hydrogen, carbon dioxide, oxygen and nitrogen. The hydrogen and nitrogen were mixed and passed to the anode inlet of the fuel cell. Carbon dioxide mixed with air entered the cathode air inlet. The gas flow apparatus was a D07 mass flow controller. The upper and lower end plates of the fuel cell were connected to an FT6800 series (Faith) Electronic Load. The discharge test of the stack was conducted by controlling a computer connected to the load. Figure 4 shows the gas flow control equipment, electronic load, and stack heating furnace in the Huaneng Clean Energy Institute (HNCERI).

Fig. 2 Molten carbonate fuel cell stack

Table 1 Molten carbonate fuel cell parameters

Molten carbonate fuel cell item	Parameter
Anode	Porous Ni
Cathode	Porous NiO
Electrode area	2000 cm^2
Matrix	α-$LiAlO_2$
Electrolyte	Li_2CO_3/K_2CO_3
Bipolar plate	316L
Sealing method	Wet seal
Number of single cells	80

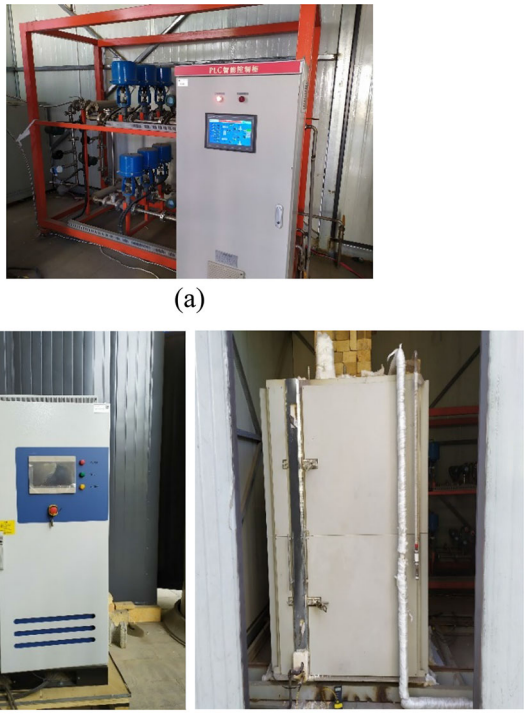

Fig. 4 Fuel cell test equipment: **a** gas flow control; **b** electronic load; **c** fuel cell stack heating furnace

3 Results and discussion

3.1 Characteristics of fuel cell materials

Figure 5 shows the morphology of the MCFC electrode. Figure 5a shows the morphology of the nickel electrode after casting; it has a loose porous structure. The nickel powder is connected by a binder, and the pore diameter of the porous structure is smaller than 10 μm. Figure 5b and c show, respectively, the morphology of the anode and

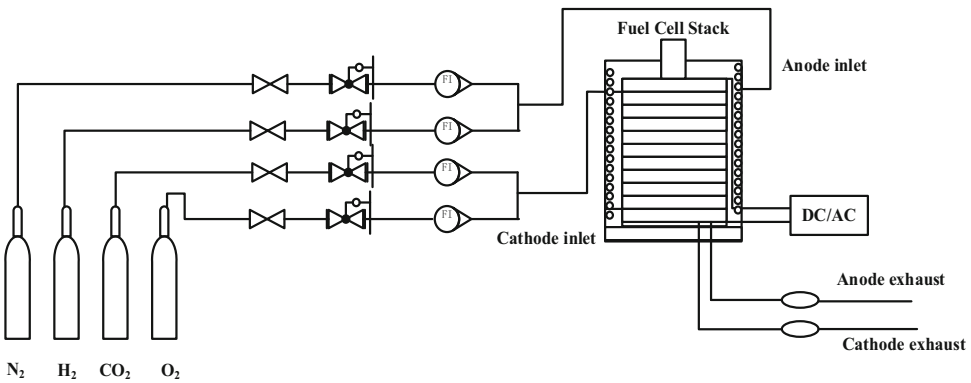

Fig. 3 Fuel cell test device

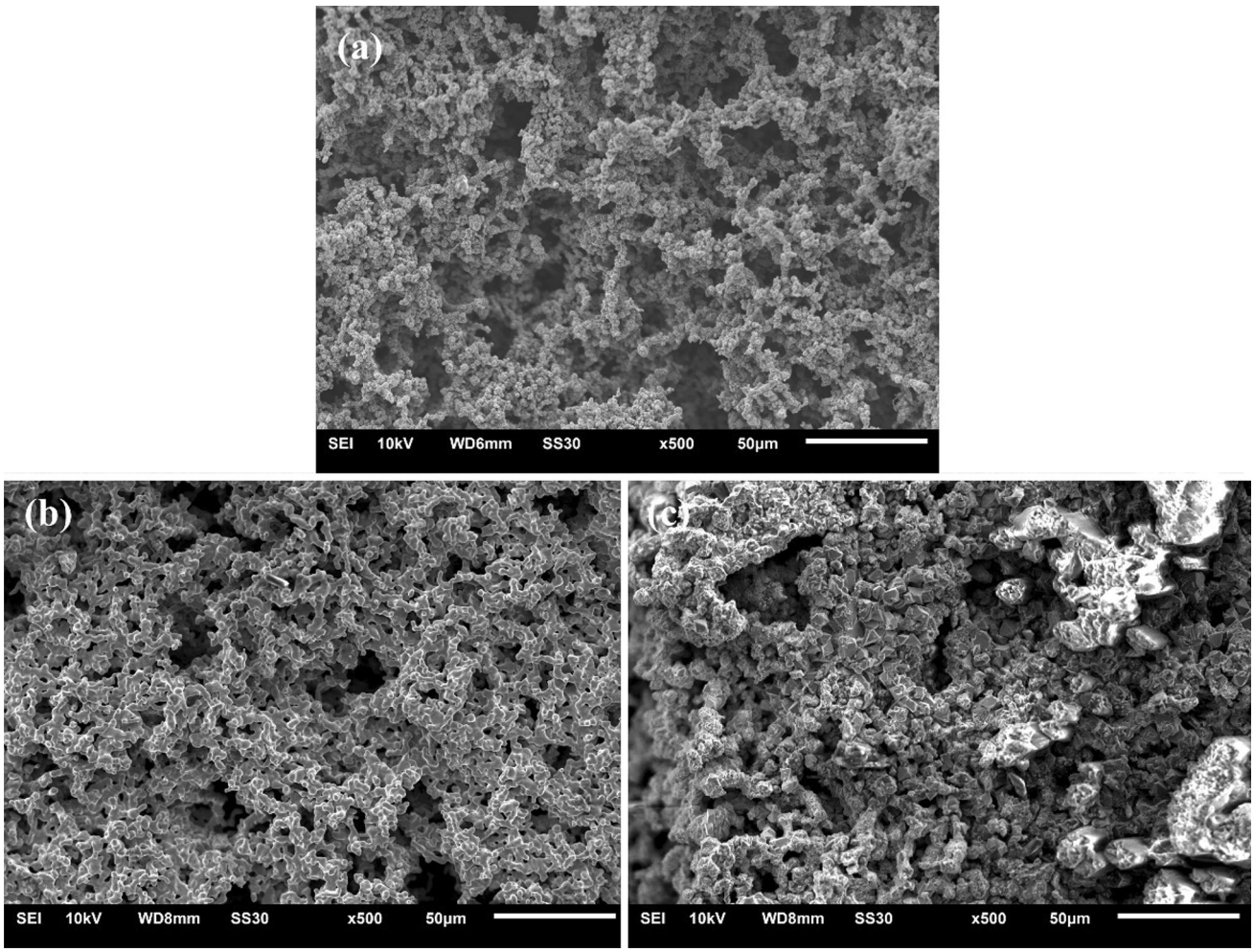

Fig. 5 **a** Electrode before firing; **b** anode morphology after firing; **c** cathode morphology after firing

cathode electrodes after firing. The electrodes are all loose-structured and porous, and the distribution of pores is relatively uniform. The average pore diameter measured is 6.5 μm.

Figure 6 shows the X-ray diffraction spectrum of the powder prepared from the separator. The main α-$LiAlO_2$ can be identified in the figure, and there is a small amount of Li_2CO_3 that did not participate in the reaction.

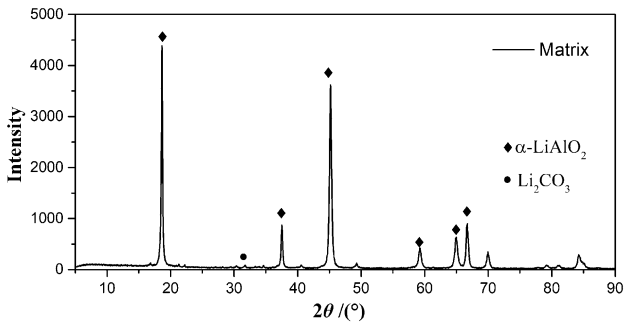

Fig. 6 X-ray diffraction spectrum of matrix powder

3.2 Test of 10 kW fuel cell power generation

The high-temperature fuel cell stack is an important part of the IGFC power generation system, with nearly zero CO_2 emissions. As shown in Fig. 7, the fuel gas enters the high-temperature fuel cell and reacts to convert chemical energy into electricity and heat. The gas produced in the reaction is discharged with the anode exhaust of the fuel cell to enter the next stage, which is the utilization of waste heat and waste gas.

The fuel cell power generation system is mainly composed of MCFCs and SOFCs. Our research team is mainly working on the MCFC power generation module and is working to provide a 100 kW MCFC stack and system for the IGFC power generation system. The 10 kW MCFC power generation unit is the smallest module in the fuel stack, which provides a research basis for the development of modules operating at 20 kW, 50 kW, and 100 kW.

The voltage–current performance of a single cell was determined before experiments on the 10 kW MCFC stack

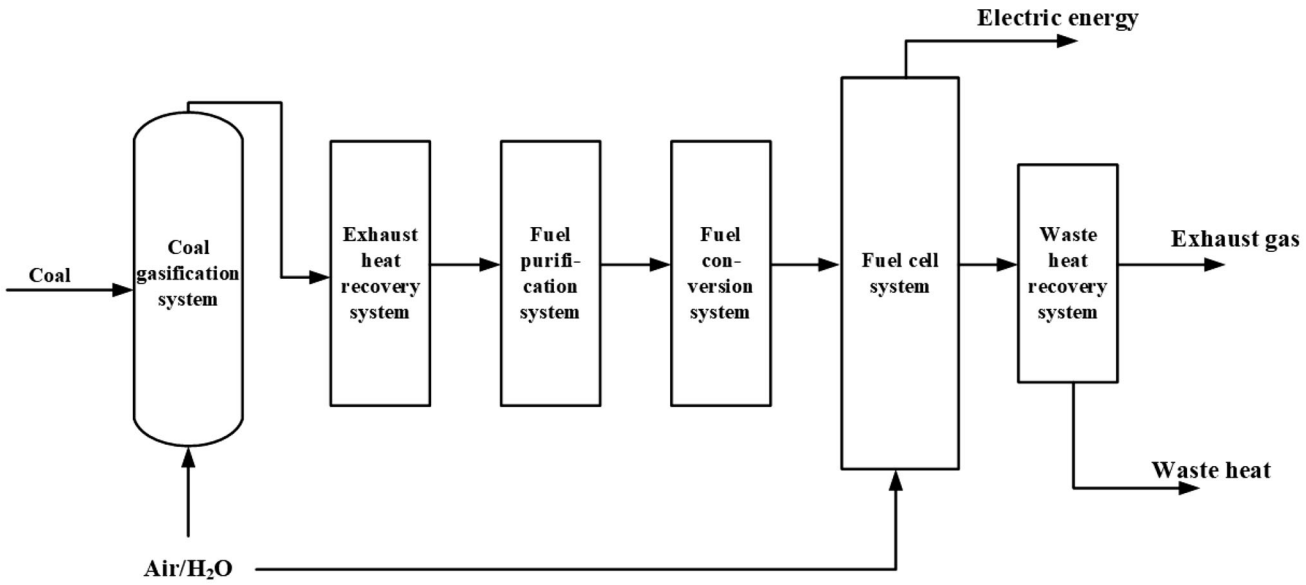

Fig. 7 IGFC power generation

were conducted. Figure 8 is a graph of voltage as a function of current density. It can be seen that the open circuit voltage of the stack is 1.23 V. The voltage continues to decrease with increasing current density. The current density is greater than 100 mA/cm^2 when the discharge voltage is 0.7 V. However, the current density reduced to below 100 mA/cm^2, becoming closer to 90 mA/cm^2, with an increase in operation time.

Table 2 gives the gas flows of the experimental stack. The power curve of an MCFC is shown in Fig. 9. The working voltage is 56 V (0.7 V, 80 single cells). It can be seen that the maximum power of the MCFC is 11.7 kW. The cell can operate at a power of greater than 10 kW for 140 min. However, the power decreases with increasing operating time. It was also found that the experimental gas

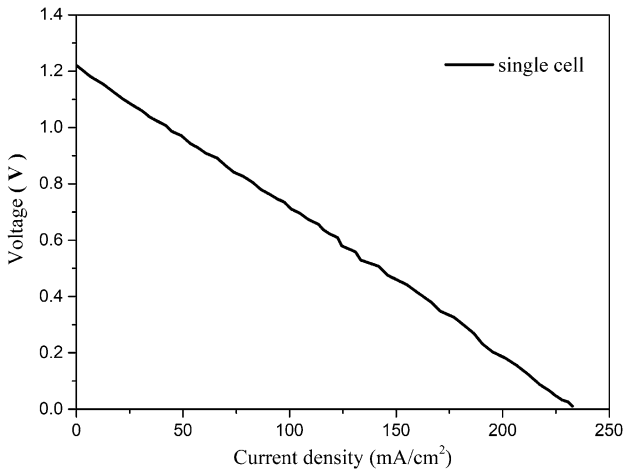

Fig. 8 Voltage as a function of current density for a single cell

Table 2 Gas flow of the experimental fuel cell

Gas flow content	Theoretical value (L/min)	Experimental value (L/min)
H_2	110	160
CO_2	110	165
O_2 (air)	55 (275)	80 (400)
N_2	30	30

flow values exceed the theoretical value. The preliminary reasons for this are the gas flow ratio and the wet seal method (Koh et al. 2000). Through the analysis of the fuel cell module, it was found that in the combined bipolar plate adopted, a sealing material was formed from the matrix and electrolyte. However, it was found that leakage occurred between the combined bipolar plates after the cooling process. By comparing with the structures of fuel cell bipolar plates created in other countries, it was found that the cathode and anode flow field were separated by welding the bipolar plates. On comparing our fuel cell structures with the structures of fuel cell bipolar plates created in other countries, it was found that, in the other fuel cell structures, the cathode and anode flow field were separated by welding the bipolar plates. This welding method can effectively reduce the possibility of fuel leakage. This is a technological gap for us, which we plan to address in future experiments. In addition, the ratio of fuel gas has some influence on the performance of the fuel cell. Concerning the difference of reaction speed and concentration polarization of anode and cathode gases in the fuel cell, the theoretical ratio of fuel gas may not deliver the theoretical

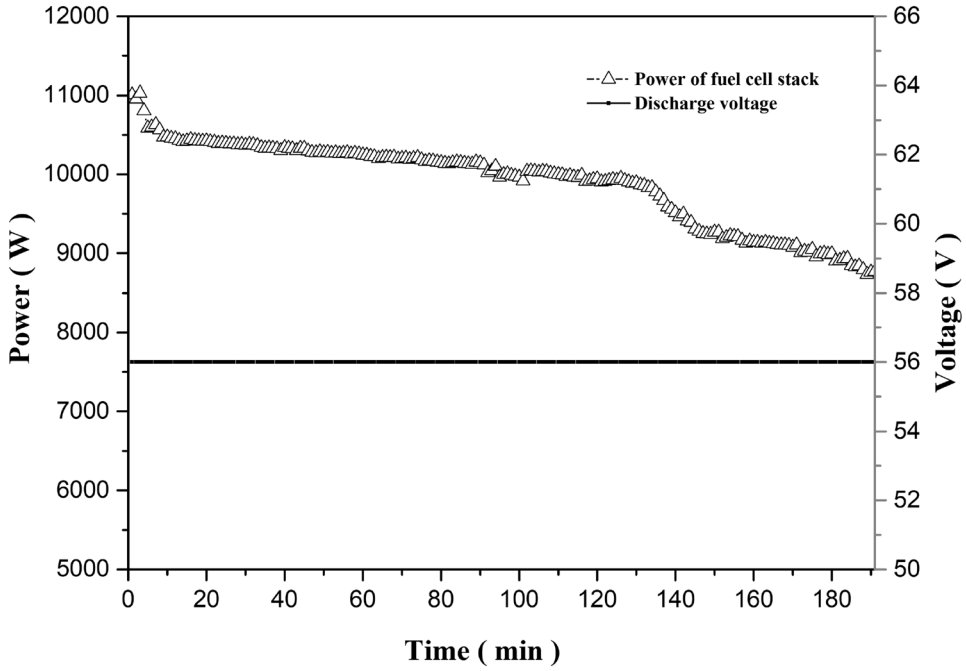

Fig. 9 Fuel cell stack operating voltage and power

power value. It is especially necessary to consider the proportion of fuel gases used.

The molar ratios of gases are H_2:N_2 (0.84:0.16) and O_2:CO_2 (0.33:0.67). The efficiency of H_2 fuel utilization can be calculated as (Yi 2003)

$$\eta_{\text{fuel}} = \frac{Pt}{UeN_{\text{A}}\rho V_{\text{H}_2}} \quad (1)$$

where, P is the fuel cell power (W); t is time (min); U is the operating voltage (V); ρ is the density of hydrogen (g/L); V_{H_2} is the hydrogen flow (L/min); e is the charge on an electron (C); and N_{A} is the Avogadro constant. The efficiency of H_2 fuel, η_{fuel}, is 73%.

3.3 Questions and challenges

In this study, the power of the MCFC achieved was 10 kW, which was required by the project. There were problems, which were caused by a fuel leak occurring during the reaction of the stack, and it was found that there was insufficient sealing of the stack. The stack inlet was analyzed. Because the gas flow increased, salt was lost between the single cells; this resulted in a decline in the wet seal strength of the fuel cell. In addition, the matching of the electrode separator used needs to be further improved, the electrode needs to be made thinner, the gas diffusion distance should be reduced, and the reaction rate should be increased. In this study, the anode and cathode were of the same thickness and porosity, which is at odds with current fuel cell electrode technology in other countries. It is necessary to continue experimental work to reduce the thickness of the cathode electrode and increase the porosity. From the analysis of current density results (> 100 mA/cm^2), the total number of designed fuel cell stacks can theoretically produce an output power of 20 kW, but in our experiments, this output power could not be attained. During the experiments, the peak power was 16.7 kW, but the stability was not ideal; this was also related to the aforementioned problems. The long-life operation of the stack is an important factor for successful fuel cell demonstration, and experiments and tests to investigate the long-term operation of fuel cells should be conducted.

The key material technology of the fuel cell in this research process has been consistent with technology in other countries. The technology gap mainly lies in mutual matching of the internal materials of single cells and assembly matching between the single cells, as well as achieving the optimum gas flow rate and flow control during fuel cell operation. In overcoming the current technical problems, it is believed that the technology gap will be reduced, in order to accelerate demonstration of the application of MCFCs in China.

4 Conclusions

In this study, a 10 kW fuel cell stack was established based on previous research. The effects of the materials and operating conditions on a 10 kW MCFC system were

analyzed. The pore size of the key MCFC electrode material was 6.5 μm and the matrix material was α-$LiAlO_2$. The experimental open circuit voltage of the single cell was 1.23 V. The current density was greater than 100 mA/cm^2 for an operating voltage of 0.7 V. The 10 kW fuel cell stack comprised 80 single fuel cells with a total area of 2000 cm^2. A fuel cell stack power of 11.7 kW could be achieved for an operating voltage of 56 V. The experimental results effectively reflected the current technology of the MCFC key materials, as well as fuel cell stack operation. The successful development of the 10 kW stack has promoted the progress of domestic MCFC technology. However, this study focused on engineering applications and ignores a theoretical analysis of the experimental process and results. Additional tests are necessary to ensure the integrity of the results. Furthermore, there were certain problems in the long-term operation of the fuel stack, such as material stability and maintaining a wet seal of the fuel cell stack.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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