

A comparative study of different carbon fuels in an electrolyte-supported hybrid direct carbon fuel cell



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HIGHLIGHTS

- Three types and three particle sizes of carbon fuels were investigated.
- High electrochemical performance has been achieved in the DCFC.
- Sulphur poisoning has detrimental effect on the DCFC performance.

ARTICLE INFO

Article history:

Received 19 October 2012

Received in revised form 22 January 2013

Accepted 22 March 2013

Available online 16 April 2013

Keywords:

Direct carbon fuel cell

Electrolyte-supported fuel cell

Solid carbon

Carbonates

ABSTRACT

An electrolyte-supported hybrid direct carbon fuel cell (DCFC) was used to evaluate the performance of different carbon fuels in this study. The direct carbon fuel cell consists of a samarium doped ceria (SDC) electrolyte, a Ni/SDC anode and a $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ cathode. Three types of carbon (graphite, coal and activated carbon) and three particle sizes of activated carbon (70, 250 and 500 μm) were investigated at 650–750 °C. The electrochemical reactivity of these three types of carbon fuels was in the order of activated carbon > German creek coal > graphite. Sulphur in German creek coal has poisoning effect on Ni catalyst resulting in lower power density of the fuel cells. The activated carbon (250 μm) fuelled hybrid DCFC achieved a peak power density of 158.3 mW cm^{-2} at 750 °C along with the maximum current density of 561.5 mA cm^{-2} . However, the stability of the hybrid DCFC is poor and need to be improved at the present.

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1. Introduction

The direct carbon fuel cell (DCFC) is a unique concept which can convert the chemical energy of carbon or biomass directly into electricity without a reformation process. Direct carbon fuel cells (DCFCs) could offer significant advantages, including near 100% thermodynamic efficiency of the carbon oxidation reaction, a high practical efficiency far superior to coal-fired generation and greater than many other fuel cell technologies, abundant solid carbonaceous fuels available such as coal and biomass, and an almost pure stream of carbon dioxide exhaust ready for capture and subsequent sequestration leading to substantial cost and energy savings [1–3]. Although DCFC is in its early stages of development, it has become a hot research topic as a potential practical energy technology. Until now, not only there have been different attempts to convert carbon materials directly into electricity in the DCFC and show potential commercialisation due to their promising performance, but also lots of updated review papers have been written on this topic [4–9]. Based on the types of electrolytes used, direct

carbon fuel cells can be mainly grouped into three categories, molten hydroxide [10–14], molten carbonate [3,6,15–21] and solid oxide electrolyte fuel cells [2,22–26]. One of the latest developments in the DCFC technology is to disperse solid carbon particulates in a molten carbonate as an extended anode at high temperatures [27–32]. For this type of DCFC (named hybrid DCFC), molten carbonates are remained at the anode, but the cathode is not exposed to carbonates, thus advanced cathode materials can be used to improve oxygen reduction reaction and the resistance of anode can be reduced as a result of the facilitated oxidation of carbon in molten carbonates. The status of these DCFCs has been summarised in Table 1.

Although various carbon fuels have been investigated using different DCFC designs, including crystal structure, particle size, surface modification and impurity content, the detailed mechanism of carbon electrochemical oxidation is still not fully understood in most DCFC designs. Furthermore, most of these researchers dispersed carbon particles in the molten carbonate as an extended anode with carbonic ions as the main active specie (Fig. 1a), and employed molten carbonates as the electrolyte [3,6,15–21]. The electrochemical reactivity of these types of carbon fuels are ranked as following: activated carbon > coal > graphite in the previous

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Table 1
Summarised status of these DCFCs.

Electrolyte	Hydroxide	Carbonate	Solid oxide	
DCFC name	MARK III-A	Tilted DCFC	Metal anode	Hybrid DCFC
Main research group	SARA	LLNL	Gorte's team CellTech (JP-8)	Irvine's team
Performance	58mW cm ⁻² for 540 h at 630 °C [10]	96mW cm ⁻² at 800 °C [5]	300mW cm ⁻² at 750 °C (molten Sb anode) [40]	36mW cm ⁻² at 700 °C under 0.7 V for 17 h [41]
Scale up	Have concept sketch for a commercial prototype	Readily scaled-up	Developing a field battery charger (20–100 W, JP-8)	Small scaled up (4 cm ²)

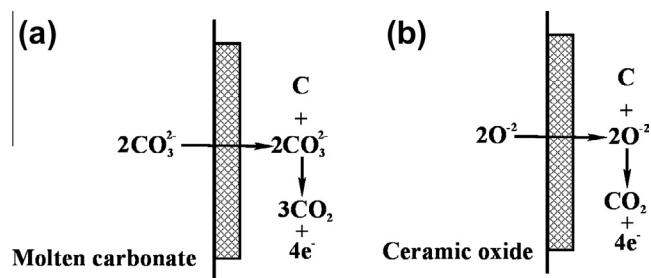


Fig. 1. Possible reaction mechanism of the direct oxidation of solid carbon in the DCFC with (a) carbonic ion as main oxygen active species and (b) oxygen ion as main oxygen active species in the carbon/carbonate slurry.

study [21,33] while our group's previous results shows a certain advantage to use GK coal over activated carbon [34]. The ambiguous results may be originating from different DCFC structure. To the best of our knowledge, no study has been conducted on the effect of carbon fuels on the performance of this hybrid structured DCFC, using oxygen ion as the main active specie (Fig. 1b). In this study, the effects of carbon fuels on the performance of the hybrid DCFC were investigated with the carbonate-solid carbon slurry in the anode compartment (Fig. 1b), including different types of carbon, different size of carbon and sulphur impurity.

2. Experimental

2.1. Fabrication of the fuel cell

The SDC electrolyte pellets with the thickness of 550–600 μm and diameter of 14 mm were prepared by dry pressing commercial SDC powder (NexTech) and sintering at 1400 °C for 5 h. The NiO power (NexTech) and SDC powder were mixed with terpineol and ethyle cellulose using mortar and pestle in the weight ratio of NiO:SDC = 6:4, and then tape casted on one surface of the SDC pellet as the anode. BSCF powders, synthesized by a combined EDTA-citrate complex process [35] were mixed with 2-propanol and glycerol using ball mill for 2 h to form BSCF ink. The BSCF ink was sprayed on the other side of SDC pellet as the cathode. The sintering temperature of the anode and cathode were 1400 °C and 1000 °C, respectively, and the geometric surface area of the sintered electrodes was 0.28 cm². Silver paste was painted on both sides as current collectors. The morphology of electrodes and electrolyte of the fuel cell was observed by a field emission scanning electron microscope (FESEM, JEOL 7001).

Three types of carbon were used in this study, including graphite (GC < 20 μm), German Creek coal (GK < 200 μm) and different sizes of the activated carbon (AC < 70 μm, 125–250 μm and 450–500 μm). A eutectic mixture of 62 mol% Li₂CO₃ and 38 mol% K₂CO₃ at a mole ratio of carbon to carbonate of 1:1 was prepared and used to fill the tube as the extended anode according to the previous results [19,32].

2.2. Cell sketch and measurement

Fig. 2 shows a schematic design of the hybrid direct carbon fuel cell, which is similar to other designs [27–32]. The silver wire was attached to electrodes using silver paste. The silver paste was sintered at 600 °C before the pellet was attached to an alumina tube (supplied by AOF ceramic) with a ceramic paste (Aremco 668). Electrochemical oxidation of solid carbon in this study was carried out under batch fuel conditions and the amount of carbon/carbonate mixture was approximately 2 g. Air and Ar (approximately 100 ml min⁻¹) was fed to the cathode and anode, respectively.

As we all know, high temperature benefit the carbon oxidation, however, high operating temperature may lead to reverse Boudouard reaction, which may decrease the fuel efficiency. Moreover, the problem of the sealing and corrosion become serious with temperature increasing. Therefore, the operating temperature was chosen in the range of 650–750 °C. The cell was heated by a top-loading Lindberg crucible furnace to the designed temperature. Electrochemical measurements were carried out with a potentiostat (Autolab PGSTAT302), using the GPES and FRA software package (Version 4.9). The measurement was started at 750 °C and the temperature was decreased at 25 °C interval to 650 °C. *I*–*V* curves measurements were started by potential sweep at a scan rate of 0.05 V s⁻¹ once a steady OCV was reached and were repeated for several times until the stable performance was achieved. Impedance spectra were carried out in the frequency range of 0.01–1 MHz.

2.3. Analytical characterisation of carbons

The commercial granular activated carbon (Calgon BPL) was ground and sieved into different sizes, and three different particle

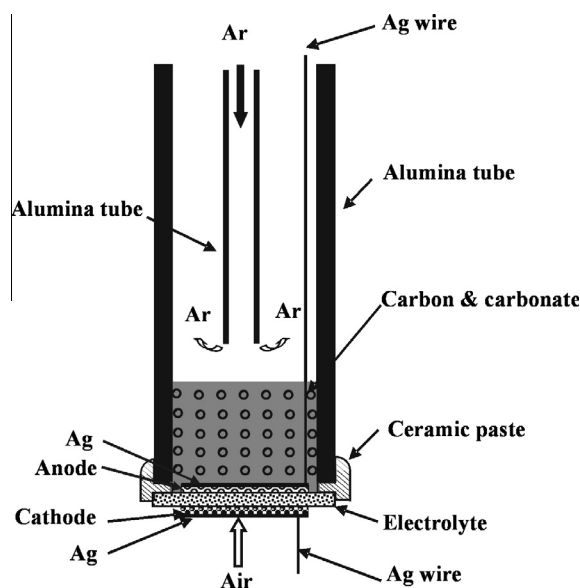


Fig. 2. Schematic of the direct carbon fuel cell.

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