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Enhancing triple-phase boundary at fuel electrode of direct carbon fuel cell using a fuel-filled ceria-coated porous anode

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ABSTRACT

A new type of high-temperature fuel cell using solid carbon as a fuel, which is called a direct carbon fuel cell (DCFC), recently attracts scientific and industrial attention due to its excellent electrochemical efficiency, less production of CO₂, and no need of CO₂ separation. However, the state-of-the-art technology on the DCFC still stays in an idea developing stage, mainly because of fuel-related difficulties: a discontinuous fuel supply and a very limited formation of triple phase boundary. In this study, we focused on how to enhance the formation of triple phase boundary at the fuel electrode: using a porous Ni anode filled with carbon particles to enhance the fuel-electrode physical contact and making the porous anode wettable by ceria coating the anode. We demonstrated for the first time that the two ideas are quite successful, leading to 700% increase in a maximal power density and 500% increase in a maximal current density with respect to the standard case.

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Introduction

A direct carbon fuel cell (DCFC) that uses carbon-rich solid materials as a fuel produces electricity while releasing CO₂ as a by-product. Despite this release of CO₂, DCFC is more environmental friendly than traditional carbon burning technologies. Due to its highest theoretical efficiency near 100%, DCFC requires less carbon to produce the same amount of electricity. Because only CO₂ is emitted, CO₂ separation from a flue gas in a conventional power station is not necessary. Also used is any form of carbon-containing materials such as carbon black, coals, biomass, and even industrial wastes [1]. Its potential advantages have led to DCFC gaining much more attention.

Many researches for the DCFCs have been performed in a way to modify the fuel electrode of the fuel cells using gaseous fuels for the use of various solid fuels. For example, a cylindrical carbon rod has been inserted in a molten electrolyte pool, in an attempt to use as a solid fuel and an anode [2–4]. Though the overall simplicity in design makes it commercially attractive, there exist a few drawbacks: a surface-limited triple-phase boundary, discontinuous (manual) refueling, and a possible oxidative (rather than electrochemical) consumption of the electrode by dissolved air. Cooper et al. [5–7] in LLNL developed a tilted DCFC system capable of continual refueling with the carbonate-carbon slurry flowing down on the anode. They have also designed a self-feeding cell incorporating pneumatic refueling and internal pyrolysis of the coal [5]. Though they resolved the issue of continuous fuel supply, the

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electrochemical reaction is constrained on the anode surface. Besides, Vutetakis et al. developed a test bed consisting of a gold working electrode in contact with a coal powder-carbonate slurry, a graphite counter electrode and a gold reference electrode [8]. Though their design enabled to monitor various gas species evolving from the electrochemical reaction [9–11], their cell also suffered from a very limited fuel/electrode/electrolyte contact.

Another type of DCFC was developed on the basis of the solid oxide fuel cell (SOFC). Typical operation temperature of this system is 800–1000 °C, higher than that of the molten carbonate (MC)-based DCFC, although there is a strong technology push to lower the temperature. Note that at such a high temperature, the carbon fuel is not only reacted electrochemically with oxygen ions at the anode ($C + 2O^{2-} = CO_2 + 4e^-$) but also gasified into CO through reverse Boudouard reaction ($C + CO_2 = 2CO$) followed by the electrochemical oxidation of CO ($2CO + 2O^{2-} = 2CO_2 + 4e^-$). While the former route requires a direct fuel/electrode/electrolyte contact to be present, the latter does not require so that carbon fuels can be supplied with fluidized or packed beds or even be in a small distance from the anode. Despite this facile fuel supply in this system, it is well known that the CO as an electrochemical reactant reduces the theoretical efficiency and fuel utilization as compared to the direct route. Moreover, the indirect route was not originally aimed in the DCFC system, but rather classified to another fuel reforming version of the SOFC.

Hence, we turned our attention back to the direct route which becomes more probable at temperatures below 700 °C where the reverse Boudouard reaction is thermodynamically inhibited. Overall, it is summarized that the current DCFC technology utilizing the direct anodic reaction of carbon to CO_2 suffers from a discontinuous fuel supply and a limited formation of triple phase boundary. Owing to the triple-phase boundary issue associated with its uneasy handling of solid

fuels, the power density of the DCFC is still much lower than that of gas-fueled MCFC or SOFC, making the relevant researches remain in an idea-developing stage or feasibility test level. Despite a large number of relevant papers, there are very few papers attempting to increase the triple phase boundary in the DCFC anode. To the best of our knowledge, only one paper [12] reported that a porous cathode was tested to enhance the cathodic reaction ($2CO_2 + O_2 + 4e^- = 2CO_3^{2-}$). However the result is not quite satisfactory, reporting a maximum power density of $\sim 30 \text{ mW cm}^{-2}$ at a current density of 50 mA cm^{-2} .

Thus, this study was focused on enhancing the triple phase boundary at the anode of a MC-based DCFC and demonstrating its effect on the power generation. A porous nickel anode filled with carbon powder was first tested in comparison with a reference case in which carbon powder was put on top of the electrode. Another porous nickel anode was coated with ceria in order to make the electrode electrolyte-wettable and to improve triple phase boundary further. Using a coin-type DCFC cell, it was demonstrated that these ideas are so effective that the electrical power of the cell was increased finally by a factor of seven.

Experimental section

Fig. 1 shows a schematic drawing of a DCFC system for evaluation of a coin-type unit cell. The coin cell comprised a porous Ni anode (INCOFOAM[®], porosity 97%), a thin-plate NiO cathode, two sheets of Pt current collectors, and a molten two-component electrolyte sandwiched with two sheets of ceramic matrices which electrically insulated the two electrodes. Detail specifications of the components are summarized in Table 1. Three different modifications of the fuel-anode assembly were considered in a way that investigated the influences of triple-phase boundary and wettability of the

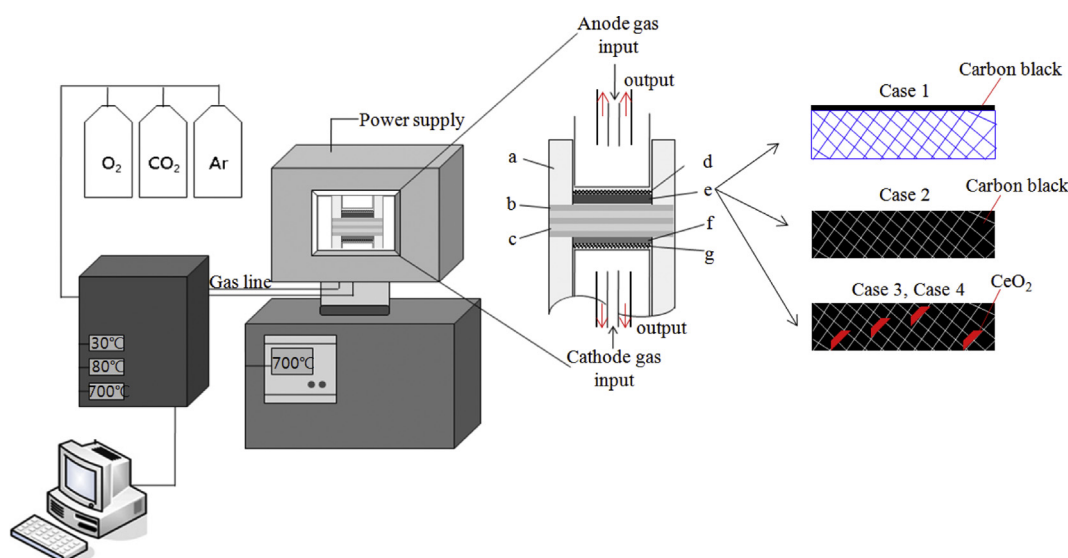


Fig. 1 – Schematics of the present direct carbon fuel cell comprising a) ceramic tube, b) matrix, c) electrolyte, d) anode current collector, e) anode, f) cathode, g) cathode current collector; case 1) putting carbon fuel on top of the porous nickel electrode, case 2) carbon fueling inside the porous nickel electrode, cases 3) and 4) coating the nickel electrode with CeO_2 at two different contents.

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