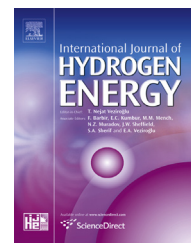


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All-solid-state direct carbon fuel cells with thin yttrium-stabilized-zirconia electrolyte supported on nickel and iron bimetal-based anodes

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ABSTRACT

Solid oxide fuel cells (SOFCs) with thin yttrium-stabilized-zirconia (YSZ) electrolyte supported on composite anode of nickel-iron bimetal and YSZ are prepared and operated directly on Fe-loaded activated carbon fuel, without any liquid medium or purging gas. The composition of the anode is represented as $\text{Ni}_{1-x}\text{Fe}_x\text{O}_6$ ($x = 0, 0.05, 0.1, 0.2, 0.3$)-YSZ. Experiment result shows that such kind of all-solid-state direct carbon solid oxide fuel cells (DC-SOFCs) perform well at 800 °C, giving maximum output power densities in the range of 425–529 mW cm^{-2} . Similar to the case of a SOFC operated on hydrogen fuel, a small amount of Fe addition into the Ni-based anode can improve the performance of a DC-SOFC and the optimum composition is $x = 0.1$. A DC-SOFC with $\text{Ni}_{0.9}\text{Fe}_{0.1}\text{O}_6$ -YSZ anode, loaded with 2.5 g Fe-loaded activated carbon fuel, is steadily operated at a constant current density of 0.1 A cm^{-2} for 15 h at 800 °C. The anodes and the DC-SOFCs are characterized through XRD, AC impedance spectroscopy, and SEM measurements. The superior performance of the Fe-added anode is analyzed accordingly.

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Introduction

With increasing concern on the global climate change caused by overconsumption of fossil fuels, there is an urgent need for reducing carbon emission. As fossil fuels, especially coal, will still be the major energy source in the foreseeable future, it is

desirable to develop novel technologies which can convert chemical energy of fossil fuels to electricity with high efficiency and low emission. Therefore, the direct carbon fuel cell (DCFC), which has been proposed since one and half centuries ago [1], has regained attention due to its high conversion efficiency and concentrated CO_2 product which can be

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sequestered with low cost [2–5]. The ideal overall electrochemical reaction in a DCFC is



The theoretical electrical conversion efficiency of this reaction is the Gibbs energy change ΔG divided by its enthalpy change ΔH . As ΔG is slightly larger than ΔH in a wide range of temperature, the conversion efficiency is over 100%, meaning some heat from external sources may be directly converted into electricity along with the DCFC operation. In a DCFC, carbon is electrochemically oxidized at the anode chamber. Compared to carbon oxidation through combustion in air, high concentrated CO_2 (without N_2) is produced. Such CO_2 can be further sequestered or utilized in a much lower cost. A report from EPRI of U.S. evaluating the feasibility of DCFC in electricity generation from coal has confirmed the advantages of DCFC over conventional coal fired power plant [6].

DCFCs can be classified into three types, according to different electrolytes: molten hydroxide [7–9], molten carbonate [10–13], and oxygen ion conducting solid oxide [14–22] among which the last one is the only solid state electrolyte. As the activation energy of carbon oxidation is high, a DCFC needs to operate at relatively high temperatures. Any liquid with high temperature is dangerous as it may cause leaking or corrosion destroying the cell system. In this respect, solid state electrolyte is superior to the liquid electrolyte. However, liquid metals [14–16] or molten salts [17–19] have been added to the anode chamber for carbon delivering in solid electrolyte DCFCs. While the liquid material helps in solving the problem of carbon refilling to DCFCs, it diminishes the merit of solid electrolyte. Gür et al. have proposed DCFCs with solid oxide electrolyte, in which coal is provided with Ar or CO_2 as carrying or reforming gas [23–25]. In recent years, our group have been focusing on developing an all-solid-state direct carbon solid oxide fuel cell (DC-SOFC), which is a SOFC operated on solid carbon directly filled in the cell as fuel, without any liquid medium or purging gas [26–34] (Fig. 1). The reaction mechanism of such DC-SOFCs was first proposed by

Nakagawa and Ishida [35] and was lately verified by Xie et al. [31]. According to the mechanism (Fig. 1a), the DC-SOFC operation is driven by the electrochemical oxidation of CO at the anode



The produced CO_2 molecules diffuse to the carbon fuel to perform the reverse Boudouard reaction



This reaction is favored at high temperatures because CO dominates the equilibrium gas composition in a C–O system that contains excess C. For example, the molar fraction of CO and CO_2 is 89% and 11%, respectively, at 800 °C. Through reaction (3), more CO is produced for reaction (2). In such way, carbon fuel is continuously delivered to the anode for the DC-SOFC operation. There is no need for chemical or even physical contact between carbon and the anode. While such DC-SOFC is still at an early stage of development and there are very few researchers reporting on it, there is an accelerated progress. In 2009, Tang et al. reported a tubular electrolyte-supporting DC-SOFC operated on graphite fuel which gave a peak power density of 9.2 mW cm^{-2} at 800 °C, but the performance dropped rapidly during a stability test [26]. Later on, by utilizing activated carbon instead of graphite as the fuel, a similar DC-SOFC had steadily operated at a current density of 12.5 mA cm^{-2} for 37 h, verifying the self-sustainability of a DC-SOFC [27]. The performance of the DC-SOFCs were significantly improved by loading Fe on activated carbon fuel and applying Ag-GDC (gadolinium doped ceria) to the anode to catalyze the Boudouard reaction and the electrochemical oxidation of CO, respectively. A peak power density of 45 mW cm^{-2} was obtained at 800 °C [28]. Meanwhile, a Ni-ScSZ anode-supported SOFC operated directly on carbon fuel was reported to produce a peak power density of 75 mW cm^{-2} at 800 °C [36]. Our group prepared a cone-shaped NiO-YSZ anode-supported three-cell-in-series stack and operated it on Fe-loaded activated carbon fuel and obtained a peak power

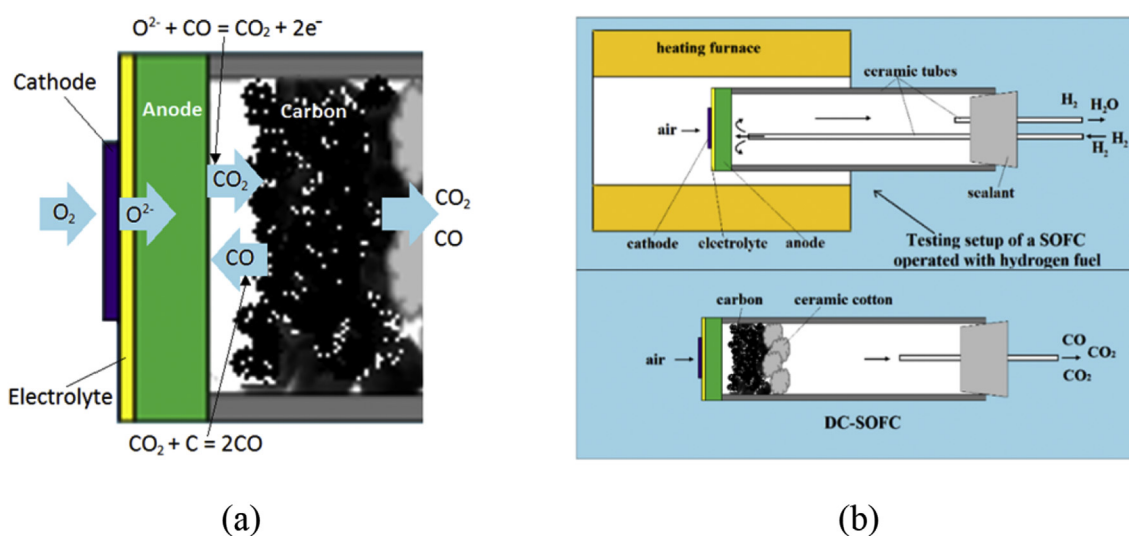


Fig. 1 – Schematic illustration of operation mechanism of a DC-SOFC (a) and testing setup of a SOFC operated on hydrogen (top) and directly on carbon (DC-SOFC) (bottom) (b).

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