Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Meticulous insight on the state of fuel in a solid oxide carbon fuel cell

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HIGHLIGHTS

• Gas chromatograms show the effect of internal gasification reactions.

Power performance is mainly governed by gas-state molecules.

• The lack of further oxidizable gases leads to decrease in performance.

• Solid carbon fuel could take part in electrochemical reactions.

ARTICLE INFO

Article history: Received 26 July 2016 Received in revised form 26 September 2016 Accepted 28 September 2016 Available online 28 September 2016

Keywords: Solid carbon Fuel cell Solid oxide electrolyte Gas-state fuel

ABSTRACT

In a solid oxide carbon fuel cell (SO-CFC), it has been widely believed that solid-state carbonaceous materials would directly take part in electrochemical reaction to generate electrons. Of late, however, it is predicted that further-oxidizable gas-state molecules, such as CO, hydrocarbons, play a crucial role in an SO-CFC operation. In this work, we controlled an anode chamber atmosphere by providing an inert purging gas to verify the role of carbon-induced gas-state molecules; that is, some experiments were carried out with a purging gas and the others were without a purging gas. Additionally, in some of the experiments, alumina wool was placed between carbon and a cell to prevent a solid-state fuel from reaching anode surface, *i.e.* alumina wool is a gas diffusion layer between a solid-state fuel and anode surface. As a result, maximum power density of 230 mW cm^{-2} was obtained when the test was taken under *no* Ar *without* alumina wool; 80 mW cm⁻² under Ar flow *without* alumina wool. In other words, it is corroborated that gas-state molecules mainly govern power performance and hence SO-CFC operation.

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1. Introduction

Carbon fuel cell (CFC) is a type of fuel cell which uses carbonaceous materials as fuel. It directly converts the chemical energy of fuel into electricity, and, hence, yields good efficiency [1]. Since it adopts the chemistry of a fuel cell, it is not subject to Carnot limitations [2]. As fuel, carbonaceous materials are beneficial, in that they are convenient to store and transport. In addition, CFC has high fuel flexibility [3–5]; that is, it can employ various kinds of carbonaceous materials, *e.g.* coal, char, biomass [6–15]. CFC can be further classified according to mechanisms and configurations of a fuel cell system [16]. Among them, especially, solid oxide carbon fuel cell (SOFC) [17]. Unlike SOFC, SO-CFC employs solid-state fuel;

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thus, direct electrochemical conversion from solid-state fuel has been widely believed as an electron-generating reaction [18].

In SO-CFCs, the chemistry of carbon and oxygen basically governs a fuel cell system; carbon is employed as fuel at an anode side, and oxygen is provided to anode from cathode *via* electrolyte as a form of oxide ion (O^{2-}). Among various possible anodic reactions, direct oxidation reactions are tabulated in Table 1. On the other hand, indirect electrochemical reactions are a series of consecutive reactions, which in turn generates electrons. In indirect electrochemical reactions, in other words, the electrochemical oxidation of chemical reaction-induced CO. Chemical reactions include partial chemical oxidation of C (C + ½ $O_2 \rightarrow$ CO) and the reverse Boudouard reaction (C + CO₂ \rightarrow 2CO).

Of late, contrary to a conventional thought in SO-CFC, some researchers have predicted gas-state molecules would play a crucial role and take part in electrochemical reactions [19–23]. They have presented evidences *via* electrochemical and/or





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 Table 1

 Major direct oxidation reactions by the chemistry of carbon-oxygen speciation.

Source	Partial oxidation		Complete oxidation	
	Chemical	Electrochemical	Chemical	Electrochemical
C CO	$C + \frac{1}{2}O_2 \rightarrow CO$	$C + O^{2-} \rightarrow CO + 2e^{-}$	$\begin{array}{l} C + O_2 \to CO_2 \\ CO + \frac{1}{2}O_2 \to CO_2 \end{array}$	$\begin{array}{c} C + 2O^{2-} \rightarrow CO_2 + 4e^- \\ CO + O^{2-} \rightarrow CO_2 + 2e^- \end{array}$

computational data, implying that the oxidation of solid carbon is not a main reaction. In addition, we also [24] previously corroborated the role of further-oxidizable gaseous molecules induced from biomass in an SO-CFC operation. In this paper, we show qualitative and electrochemical analyses to demonstrate the role of gas- and solid-state molecules within an anode chamber of SO-CFC.

2. Experimental

Experiments were carried out to identify which state of fuel (gas- or solid-state) predominantly contributes to fuel cell performance. Firstly, three electrochemical tests with graphene nanoplatelet C750 (GNP) as fuel were conducted to verify the contribution of gas-state fuels. Two of them were undergone without an inert purging gas (Ar) to an anode chamber, while the other was with Ar gas to minimize the influence of gas-state molecules at triple phase boundaries (TPBs). One of the tests without Ar was performed with alumina wool (Fig. 1) which was placed between fuel and a current collector on anode surface to prevent solid-state fuel from reaching anode surface.

For these three tests, current density-potential and -power (j-V-P) tests were carried out, using an optimally designed SO-CFC

apparatus at 900 °C. For *j*-*V*-*P* tests, an apparatus was stepfunctionally programmed as such: Hold 5 min at constant current mode for each step; Start from 0 mA cm⁻²; Add 50 mA cm⁻² to proceed to the next step after 5 min of holding; Halt when cell potential value reaches to zero. Meanwhile, gas mass chromatograms were obtained with MS (5975C, Agilent Technologies). In addition, when alumina wool was placed, Ar gas was provided at the flow rate of 50 cm³ min⁻¹ (standard condition) to hinder gasstate molecules from being diffused and reaching anode surface.

Secondly, the *j*-*V*-*P* tests to identify the contribution of solidstate fuel were additionally carried out. Fuel selection process to find an adequate carbon source was preceded the electrochemical tests. Candidates were GNP, carbon black powder (Timcal Ensaco 350G, T3) and carbon nanotube (CNT). All of these, each other, have distinctly different morphology. Among these carbons, CNT was chosen for the tests because it showed a sluggish behavior to restore cell potential from an ohmic drop. The *j*-*V*-*P* tests were performed as a function of time and gas mass chromatograms were also attained before and after the tests.

In all electrochemical experiments, the same experimental setup was employed as follows. A membrane electrode assembly (MEA) – a commercially available anode-supported button-type



Fig. 1. Images of experimental cell set-up: (a) the photograph of a sealant-attached cell with alumina (Al₂O₃) wool; (b) the photograph of a cell with carbon fuel; (c) the crosssectional schematic diagram of (b). A current collector (including wire) is not explained in (c) to enhance clarity. The wires in pictures (a) and (b) are Pt (0.5 mm in diameter), later being connected to an experimental apparatus, conducting electric current.

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