



Effect of reverse Boudouard reaction catalyst on the performance of solid oxide carbon fuel cells integrated with a dry gasifier



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ABSTRACT

A solid oxide carbon fuel cell (SO-CFC) integrated with a dry gasifier was operated on activated carbon fuel and the effect of adding a reverse Boudouard gasification catalyst on the performance and long-term operation characteristics of the SO-CFC was investigated. The reactivity of the carbon fuels for the Boudouard gasification reaction was analyzed by a thermal analysis at various operating conditions. The SO-CFC was then operated on gasified fuel gas consisting of CO₂ and CO obtained from the integrated dry gasifier. The SO-CFC operated on activated carbon fuel with 5 wt.% K₂CO₃ achieved a maximum power density of 202, 262, and 271 mW/cm² at 750, 800, and 850 °C, respectively; the SO-CFC fueled with activated carbon fuel without a catalyst meanwhile yielded maximum power density of 168 mW/cm² at 850 °C. By using electrochemical impedance spectroscopy, the effect of adding the catalyst on the gasification products and subsequently on the performance of the SO-CFC was studied. A long-term degradation test was conducted by continuously operating the SO-CFC at 50 mA/cm² for 518 h at 750 °C. During the long-term degradation test, the average degradation rate of the SO-CFC was found to be 183 mV/kh. The post-mortem SEM and XRD analyses of the SO-CFC after the long-term test revealed the presence of carbon deposits and oxidation of Ni at the anode, causing a relatively higher degree of degradation in the SO-CFC integrated with the dry gasifier during the long-term operation. The addition of the K₂CO₃ based dry gasification catalyst significantly enhances the performance of the SO-CFC integrated with dry gasification, but during long-term operation, the degradation rate is found to be higher than that of conventional H₂ fueled SOFCs.

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1. Introduction

Coal is the most abundant and inexpensive fossil fuel and currently provides approximately 30% of the primary energy of the world and more than 40% of the global electrical power generation [1]. According to estimates by the IEA, a global increase in coal-fired power generation is expected during the next few decades owing to the rapid industrialization of developing nations [2]. Clean coal technologies with higher system efficiency and better CO₂ capture possibilities, therefore, should be developed in the coming decades to mitigate the harmful emissions causing climate change and other related problems [3,4]. The direct carbon fuel cell

(DCFC) system is a promising candidate for highly-efficient coal power production [5]. This system electrochemically converts the chemical energy stored in the carbonaceous fuel into electrical energy and releases a concentrated CO₂ stream readily available for capture and sequestration [6,7]. In a solid oxide electrolyte carbon fuel cell system (SO-CFC), a carbon-based fuel is electrochemically oxidized by the delivery of oxygen ions through an oxide ion conducting electrolyte to the carbon in direct contact with the anode, as delineated in Eqs. (1) and (2):



In a SO-CFC, gaseous species such as CO are also utilized, exploiting the reaction given in Eq. (3):



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Many researchers including Nakagawa et al. and Gur et al. utilized solid carbon in direct contact with the anode and reported reasonable performance of the SO-CFC [8,9]. However, in the direct contact configuration of the SO-CFC, the direct oxidation of the carbon fuel requires a good solid-solid contact between the solid carbon fuel and the anode to increase reaction sites for carbon oxidation [10]. The solid-solid contact in the SO-CFC is achieved by using a specific clamping mechanism or by fluidization of the carbon fuel [11]. Scaling up an SO-CFC with a direct contact configuration to an operational system having a continuous solid fuel delivery, however, poses a serious design challenge. In comparison with direct carbon oxidation in the SO-CFC, carbon gasification reactions occurring in a physically separate compartment upstream of the anode in a gasifier integrated SO-CFC offers the advantage of better reactivity of the active gaseous species at the anode [12,13]. The gasifier integrated SO-CFC system also allows the heat produced by fuel cell electrochemistry to be transferred back to the endothermic gasification reactions [14]. Therefore, the SO-CFC integrated with a gasifier could feasibly utilize carbon dioxide or steam as gasifying media and convert the solid carbon fuel into CO and/or H₂.

In the recent past, many researchers have focused on developing gasifier integrated solid oxide carbon fuel cells (SO-CFCs) [15]. Lim et al. [16] reported that a 4-cell short stack SO-CFC with an integrated dry gasifier produced a maximum power of 29.4 W at 800 °C. They also showed that the SO-CFC short-stack has stable performance during continuous operation. Mushtaq et al. [17] developed and operated a 100 W class SO-CFC stack fueled by syngas obtained from coupled dry gasification of activated carbon using CO₂ as the gasifying media. Similarly, Naqvi et al. [12] utilized activated carbon fuel in an integrated steam based gasifier and obtained maximum power density of 181 mW/cm². It was reported that the addition of a K₂CO₃ gasification catalyst improves the performance of a SO-CFC integrated with a steam gasifier by affecting the steam gasification reactions. Long-term performance degradation of the SO-CFC operating on steam gasified fuels was also reported [18]. However, during the dry gasification of the carbon fuel, the conversion of carbon based fuels to CO-rich syngas is accomplished by the Boudouard gasification reaction ($\text{CO}_{2(g)} + \text{C}_{(s)} \rightleftharpoons 2\text{CO}_{(g)}$).

At higher operation temperatures (600–900 °C), the Boudouard gasification of the carbon fuel in the integrated gasifier is influenced by the type of fuel and the inorganic content inherently present in coals [19]. Among these inorganic species, alkali and alkaline earth metal carbonates exhibit catalytic activity at operation temperatures of around 900 °C [20,21]. While there is substantial information in the literature on gasification reaction mechanisms, studies on specific fuel-based catalysis in SO-CFC systems and the effect on fuel cell performance are very limited [22–24]. Gong and Huang [25] investigated the feasibility of a tubular SOFC operating on fuel gasified from a biomass-based carbon bed. They reported that coconut shell carbon with Fe₂O₃ and K₂CO₃ showed higher power density compared to the samples without a catalyst. The improved cell performance was attributed to the carbon conversion rate promoted by the catalysts. Yu et al. [26] also added K₂CO₃ catalyst to speed up the dry gasification reaction in a direct carbon fuel cell (DCFC); a fivefold increment in the maximum power density was observed at 700–850 °C. Many studies recently reported the effect of different fuels and catalysts on the performance of SO-CFCs [27–29,26,30]. Therefore, while the addition of a catalyst clearly improves the performance of the SO-CFC, it is necessary to analyze the effect of adding catalysts and different gasification parameters (temperature, CO₂ flow rate, etc.) on the electrochemical performance of gasifier integrated SO-CFCs.

This paper investigates in detail the effects of adding a K₂CO₃ catalyst along with the carbon fuel in an integrated gasifier on the electrochemical performance and long-term operation of the

SO-CFC. The Boudouard reactivity of the fuel and the electrochemical performance of an activated carbon fueled SO-CFC is systematically studied with and without a K₂CO₃ catalyst. The relative activities of these catalysts under a CO₂ atmosphere are investigated via a TGA analysis and have been related to their performance in SO-CFCs. A continuous fuel feed arrangement of solid carbon to the SO-CFC integrated gasifier is an engineering challenge for the scale-up of SO-CFC technology [31]. For a long-term degradation test, a mechanism to refill the carbon fuel in the integrated dry gasifier without interrupting the operation of the SO-CFC is devised. The SO-CFC is continuously operated for 512 h maintaining the supply of fuel and the long-term degradation rates of the SO-CFC are identified by galvanostatic tests and post-mortem material characterization.

2. Experimental

2.1. Fabrication of single cells for SO-CFC

A tubular anode supported solid oxide-based fuel cell was used. For the fabrication of a porous anode support, the proper weight of NiO and 8 mol% Y₂O₃-stabilized-ZrO₂ (8YSZ) powders were well mixed and then a pore-forming agent (carbon black, 5 wt.%) was added. Distilled water, an organic binder, a plasticizer, and a lubricant were added to the mixed powders and kneaded. The porous anode support was then extruded with an outer diameter of 13 mm. The extruded ceramic support was calcined via three stages of increasing temperature (350–750–1100 °C) and the additives (the organic binder, distilled water, plasticizer, and lubricant) were removed from the support structure by using a high-temperature treatment. On the pre-sintered porous anode support, an anode functional layer support (Ni-YSZ), an electrolyte (8YSZ), and a cathode (20 wt.% of LSM (La_{0.85}Sr_{0.15}MnO₃)/YSZ, LSM, LSCF (La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O₃) were sequentially coated and then sintered in an air condition at 1400 °C and 1150 °C, respectively. The anode functional layer (AFL) and the cathode layer were fabricated by using the slurry dip coating method and the electrolyte layer was manufactured by using the vacuum slurry coating method. In the present study, an active electrode surface area with a 10 cm² unit cell was used as a standard unit. Fig. 1(b) shows a SEM image of a cross-sectional area of a SO-CFC consisting of several layers comprising the anode, electrolyte, and cathode. The layer thickness of the AFL was 13 μm, the thickness of the electrolyte layer was 9 μm, and the cathode layer was measured as 20 μm. Ni wire (diameter 1 mm) welded on Ni felt was used as an anode current collector and Ag wire and a mesh was used as a cathode current collector on the LSCo (La_{0.6}Sr_{0.4}CoO₃, PRAXAIR) paste. The SO-CFC cell was sealed with a ceramic seal (Cerambond™, Aremco Products, Inc.). The detailed fabrication process for the tubular anode support, including information about the raw powder and the thermal treatment profile, was reported in [32].

2.2. SO-CFC performance evaluation

Fig. 1(a) shows a schematic diagram of the SO-CFC integrated with a dry gasifier used to investigate the electrochemical performance characteristics. After the installation of the SO-CFC cell in the evaluation furnace, the air chamber was placed at the bottom of the SO-CFC cell for supplying air to the cathode. A fuel chamber filled with mixed fuel (activated carbon + K₂CO₃) was connected the SO-CFC cell and CO₂ was supplied into the fuel chamber to induce the dry gasification reaction in the SO-CFC. The open circuit voltage (OCV) was observed during the temperature increase until the operating conditions were achieved, and then the polarization curves were measured in a range of 700–850 °C. The

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