



Performance evaluation of solid oxide carbon fuel cells operating on steam gasified carbon fuels



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HIGHLIGHTS

- Solid oxide carbon fuel cell was operated on different steam gasified carbon fuels.
- Among the all tested fuels, K₂CO₃ catalyzed activated carbon gave the maximum performance.
- Long-term galvanostatic test for 100 h showed relatively stable operation.

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ABSTRACT

We investigated the operating characteristics of solid oxide carbon fuel cells (SO-CFCs) integrated with a steam gasifier that used carbonaceous fuels, including activated carbon and biomass driven charcoal. Steam gasification was carried out in a specially designed gasifier, which was directly integrated with a solid-oxide based carbon fuel cell. We studied the effects of gasification temperature, steam flow rate and catalyst addition on the electrochemical performance of SO-CFC, and the results showed that among the three tested fuels, activated carbon with a K₂CO₃ catalyst performed the best. At 850 °C, maximum power density values of 108 mW/cm², 161 mW/cm² and 181 mW/cm² were achieved when the SO-CFC was operated using activated carbon, biomass driven charcoal and activated carbon with a K₂CO₃ catalyst, respectively. The SO-CFC operated continuously for 100 h and it showed relatively stable performance. This study suggests that by using a catalytic steam gasifier integrated with the SO-CFC, solid carbon fuel resources can be used for power generation with higher efficiency and minimal carbon footprint.

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1. Introduction

The dependence of the global energy economy and electricity generation on coal is primarily due to the material's abundance, reliability and low cost. Currently, more than 40% of the world's electricity is produced using coal based fuels; a recent report published by the US Energy Information Administration projects that by 2040 coal consumption will increase by nearly 50% above its current level [1]. Despite its important role as an economical and reliable energy resource, use of coal faces numerous challenges due to the material's detrimental effects on the environment,

including a huge carbon footprint and pollution caused by contaminants. To reduce the CO₂ emissions from the stack gas of coal power plants, clean coal technologies are being developed that use expensive and energy intensive post-combustion separation CO₂ capture and sequestration processes. Hence, there is a critical need to develop advanced coal power generation technologies that promise higher conversion efficiencies than the current processes, have fewer emissions and produce capture-ready CO₂ [1].

Carbon fuel cells (CFCs), which efficiently convert the chemical energy of carbon fuel into electricity by electrochemical reactions, are being widely investigated due to the increasing need for efficient and clean power generation from coal and other carbon-containing fuels (coke, biomass, waste materials, etc.). Power generation systems based on CFC technology offer higher conversion efficiency (more than 50%) than that of conventional coal-fired power plants, which typically operate at 30–35% efficiency [2]. Carbon fuel cells have an added advantage of consuming abundant

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and relatively cheap fuels derived from solid-based carbonaceous materials. Moreover, by using CFCs, hazardous emissions of carbon combustion associated gaseous products such as NO_x and SO_x can be significantly reduced, and the concentrated CO_2 product stream ensures more economical carbon capturing and sequestration [3]. Thus, CFC technology promises the conversion of carbonaceous solid fuels, including coal and biomass, etc., directly into electricity in a highly efficient and environmentally friendly manner.

A CFC consists of an ionically conducting but electronically insulating electrolyte that selectively transports an ionic form of oxygen, such as O^{2-} , OH^- or CO_3^{2-} , to a fuel electrode, where the ion electrochemically reacts with carbon and hydrogen in the fuel to form CO_2 and H_2O or only CO_2 . CFCs are classified according to the electrolyte, i.e., molten hydroxide, molten carbonate or solid oxide. Solid oxide based carbon fuel cells (SO-CFCs) with oxide ion (O^{2-}) conducting ceramics (e.g., yttria stabilized zirconia, YSZ) offer high temperature stability and improved electrochemical reactivity and do not suffer from liquid electrolyte consumption or corrosion problems [3]. When solid carbon is used as a fuel, an SO-CFC can achieve 100% theoretical efficiency. Therefore, development of solid carbon fueled SO-CFCs has recently attracted the interest of many researchers. However, the direct utilization of solid carbon fuels by the SO-CFC is technically challenging. The slow kinetics reaction between solid carbon particles and the electrolyte/electrode interface severely limits the power generation capability of the SO-CFC [4].

One solution to overcome these problems is to convert the solid carbons into SO-CFC-friendly gaseous fuels (syngas) before they enter the SO-CFC [5]. The gasification of solid carbon to convert it into syngas can be performed in an external gasifier and the syngas can be fed to the fuel cell [6,7]; or, the SO-CFC can be coupled directly to the gasifier [8,9]. Our research group has conducted detailed studies on internal gasifier integrated tubular SO-CFC stack development [10,11]. Yun et al. [10], used carbon black with molten carbonate as a fuel; an electrochemical mediator that filled in the inner part of the tubular anode supported the solid oxide fuel cell. The tubular direct carbon fuel cell (DCFC) in carbon + carbonate (1:1 wt%) fuel and air at 900 °C had a maximum power density of 122 mW/cm². The molten carbonate based fuel cell encountered several problems, including carbonate media handling and stability of the anode of the fuel cell. Therefore, the focus of research shifted toward using carbon fuel without molten carbon. For this purpose, dry gasification by carbon dioxide was used to produce syngas that was utilized as fuel for the high temperature solid oxide fuel cell. Based on this dry gasification concept, a short stack was developed by Lim et al. [11]. The 4-cell short stack of the CFC with a dry gasifier had a maximum power of 29.4 W at 800 °C. The researchers also showed that the operating behavior of a tubular CFC with basic operating variables has stable performance during continuous operation tests. Recently, Mushtaq et al. [12] developed and operated a 100 W class CFC stack fueled by syngas obtained from coupled dry gasification of activated carbon using CO_2 as the gasifying media. The 100 W stack operated for 200 h with almost stable performance.

CO_2 or steam can both be used as gasification media for the conversion of solid carbon fuels into syngas before syngas is fed to the SO-CFC [13]. When CO_2 is used as a gasifying media in a dry gasifier, it produces a mixture of CO and CO_2 from solid carbon at high temperatures (more than 750 °C) by employing the reverse Boudouard reaction [14]. CO is available in higher concentration in dry gasification because carbon is oxidized by CO_2 to form carbon monoxide in a reverse Boudouard reaction. On the other hand, steam gasification involves a complex steam-carbon reaction and water gas shift reactions to produce an H_2 rich syngas [15,16]. The dominant species in syngas from steam gasification are H_2 and CO, while only a small amount of non-reactive CO_2 is pro-

duced. When dry gasification using CO as the gasifying media is utilized, the CO is the only electrochemically active component in the gasified fuel gas. The SO-CFC operating on fuel gas from CO dry gasification produces lower open circuit voltage (OCV) below 1.0 V and a maximum power density lower than that which can be obtained when using hydrogen mixed fuels for the SO-CFC [9]. Moreover, the redox potential of dry gasification syngas ($\text{CO} + \text{CO}_2$) is significantly lower than that of hydrogen rich steam gasified syngas ($\text{CO} + \text{H}_2$), and the redox-cycle at the anode can be more detrimental for the Ni catalyst in a dry gasification fed SO-CFC [17].

Various reports in the literature also discuss the effect of the addition of a gasification catalyst in the carbonaceous fuel on the performance of the CFC. Gong et al. [5] investigated the feasibility of a tubular SOFC operating on the fuel gasified from the biomass based carbon-bed. They reported that coconut shell carbon with Fe_2O_3 and K_2CO_3 show higher power density compared to the samples without a catalyst without a catalyst. The improved cell performance was ascribed to a carbon conversion rate promoted by the catalysts. Yu et al. [18] added K_2CO_3 catalyst to speed up the dry gasification reaction in a direct carbon fuel cell (DCFC); a five-fold increment in the maximum power density was observed at 700–850 °C. Similarly, Lee et al. [19] studied the performance of a hybrid DCFC using a mixture of Li_2CO_3 – K_2CO_3 with carbon fuel as gasification catalyst and electrochemical reaction promoter. It was found that a eutectic mixture of Li_2CO_3 – K_2CO_3 produces a higher amount of CO and improves the DCFC performance. Li et al. [20] experimentally investigated the effects of catalytic gasification on the solid oxide electrolyte DCFC (direct carbon fuel cell) performance using K, Ca and Ni as catalyst in carbon black fuel. Therefore, it is pertinent that the addition of a catalyst improves the performance of the SO-CFC; however, there is a need to systematically investigate the effect of different gasification parameters (temperature, steam flow rate, etc.) on the electrochemical performance of integrated SO-CFCs.

In this study, to assess the suitability of the technology in terms of fabrication, fuel feed, operating parameters and long-term degradation, an anode supported tubular solid oxide carbon fuel cell is tested with different solid-carbon based fuels fed to an integrated steam gasifier. Based on our experimental results, we fabricated and analyzed the basic properties of the SO-CFC system coupled with steam gasification and evaluated its electrochemical performance characteristics.

2. Experimental

2.1. SO-CFC fabrication

An extruded porous tubular tube based on Ni-8 mol.% yttria-stabilized ZrO_2 (Ni-YSZ) served as an anode support for the tubular SO-CFC. The detailed fabrication process for the tubular anode support, including information about the raw powder and the thermal treatment profile, were reported in [21]. The other cell components were fabricated in thin layers and attached on the surface of the tubular support. The powders of the tubular anode support and activated carbon as a pore former were weighed and mixed in ethanol by ball milling, and then dried. An organic binder and distilled water were added to the dried powder; then, a well-dispersed paste was extruded in the form of a tubular anode support. The extruded tubes were dried in a rolling dryer at room temperature and then pre-sintered at 1100 °C. After that, the dip-coating method was used to deposit an anode functional layer (AFL) onto the pre-sintered anode support to form a homogeneous surface for coating the electrolyte film. To form a thin, crack-free layer, a YSZ electrolyte layer was coated onto the surface of the

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