



# Long-term performance degradation study of solid oxide carbon fuel cells integrated with a steam gasifier



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## ABSTRACT

We investigated the long-term performance degradation in a solid oxide carbon fuel cell (SO-CFC) integrated with a steam gasifier that used activated carbon as a fuel. The steam gasifier-integrated SO-CFC was continuously operated for 2000 h at 750 °C under galvanostatic conditions. Fuel analysis, in-situ electrochemical tests, and post-mortem characterization were performed to determine the dominant degradation factors during the extended operation of the SO-CFC. During the 2000 h continuous operation, the overall degradation rate of the SO-CFC was 0.108 V per 1000 h. Electrochemical impedance spectroscopy analysis showed that the polarization and ohmic resistance of the SO-CFC increased gradually with the operation time. The post-mortem diagnosis via SEM, XRD, EDS, and XPS analyses revealed that the increased Ni particle coarsening and carbon deposition at the anode functional layer induced increased performance degradation in the SO-CFC. The long-term durability study of the SO-CFC with a steam gasifier confirmed that conventional anode materials result in rapid performance degradation during 2000 h continuous operation, and further development of anode materials will be necessary for the long and durable operation of SO-CFCs.

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## 1. Introduction

Coal is the second largest primary energy source in the world after oil, and it is the major fuel in thermal power plants due to its abundance and low price. Currently, in the United States, more than 50% of electricity is produced from coal-based power plants, while in China and India it accounts for more than 70% of electricity production [1]. However, conventional coal-fired power plants have relatively low efficiencies (<40%) and produce a considerable amount of greenhouse gas emissions (800–900 g/kWh) [2,3], which are major triggers of climate change. Therefore, improvements in clean coal utilization technologies must continue in order to achieve higher conversion efficiencies and lower CO<sub>2</sub> emissions

from the stack gas of power plants, and to produce capture-ready CO<sub>2</sub> for an efficient carbon capture and sequestration process [4].

Carbon fuel cells (CFC's), which electrochemically convert the chemical energy of carbon fuel into electricity without combustion, 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.) [5–7]. Among the different types of carbon fuel cells, solid oxide-based carbon fuel cells (SO-CFC) with oxide ion (O<sup>2-</sup>) conducting ceramics (e.g., yttria-stabilized zirconia, YSZ) have been proposed as the most promising option due to their simple construction, improved electrochemical reactivity, and fewer maintenance requirements [8]. Using solid oxide based fuel cells, Nakagawa et al. Gur et al. and many other researchers utilized solid carbon fuel as a direct contact with the anode and reported a reasonable performance of direct carbon fuel cells (DCFC) [9,10]. However, the direct utilization of solid carbon fuels in the SO-CFC is technically challenging. The major issue with CF-SOFCs currently is the difficulty of using the porous anodes to transport the solid fuel into the anode

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reaction zone or to the triple phase boundaries (TPBs) [10]. Other challenges include fuel processing and fuel quality requirements, and corrosion of cell components, particularly when molten salts are used as the fuel carrier [11]. Therefore, the scaling up of the DCFC to an operational system with a continuous solid fuel delivery poses a serious design challenge. In comparison with the DCFC, carbon gasification reactions occur in a physically separate compartment upstream of the anode in an integrated CFC [7]. In the recent past, many researchers have focused on the development of gasifier integrated solid oxide carbon fuel cell (SO-CFC) [12,13]. Lim et al. [14] reported that a 4-cell short stack of the 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. [15] developed and operated a 100 W class SO-CFC stack fueled by syngas obtained from a coupled dry gasification of activated carbon using CO<sub>2</sub> as the gasifying media. Similarly, Naqvi et al. [16] utilized activated carbon fuel in an integrated steam based gasifier and obtained a maximum power density of 181 mWcm<sup>-2</sup>. It was reported that the addition of a K<sub>2</sub>CO<sub>3</sub> gasification catalyst improved the performance of the SO-CFC integrated with a steam gasifier by affecting the steam gasification reactions.

Steam gasification involves a complex steam-carbon reaction and water gas shift reactions that produce an H<sub>2</sub>-rich gasified fuel gas [17]. The dominant species in syngas from steam gasification is H<sub>2</sub>, while only a small amount of non-reactive CO<sub>2</sub> is produced. Gür et al. [18] studied the performance of fluidized-bed SO-CFCs using a Ni/YSZ cermet, anode-supported, tubular SO-CFC at 850 °C with CO<sub>2</sub> as the fluidizing gas. A fluidized bed of Alaska coal fuel was gasified at 930 °C by flowing CO<sub>2</sub> to generate CO. The highest cell power density achieved was 450 mWcm<sup>-2</sup> at 0.64 V with a 35.7% electrical conversion efficiency based on CO utilization. After performance tests, the SOFC anodes were visually examined, and no significant signs of carbon deposition (i.e., coking) were observed. Independent tests in similar cells performed under galvanostatic conditions in flowing pure CO also indicated long-term stability of these anode-supported SOFC cells, with no sign of degradation over several hundred hours of operation [19]. In our recently published paper [16], the steam gasifier integrated SO-CFC was operated for an initial 100 h and stable performance was reported. However, to our knowledge, no study has been made on the long-term (>100 h) degradation of SO-CFCs operating on steam gasified carbon fuels.

As the state-of-the-art anode supported SO-CFC uses nickel-yttria stabilized zirconia (Ni-YSZ) as anode material, the Ni catalyst in the SO-CFC anode poses several challenges related to stability and long-term operation when it is exposed to SO-CFC operating conditions. In the presence of steam gasified syngas fuel, the Ni catalyst becomes vulnerable to carbon coking and deposition [20]. This carbon coking phenomenon results in activity loss of the Ni catalyst, and the performance of the SO-CFC is significantly decreased. The presence of a significant amount of sulfur in almost all coals also causes poisoning of the Ni catalyst [21]. When these two degradation processes, i.e., the carbon coking and sulfur poisoning, are combined with the fundamental SOFC degradation phenomena, including Ni particle agglomeration at the anode, the stability of the SO-CFC for commercial applications becomes uncertain [8]. However, there is a lack of information on performance degradation of SO-CFCs using gasified fuel gas derived from the gasification of carbon fuel in an integrated gasifier. Here, we present results of long-term degradation testing of the performance of an SO-CFC integrated with a steam gasifier during a period of 2000 h. In-situ electrochemical and post-mortem material diagnostic analysis were also performed to elucidate possible causes of the degradation in the performance of the SO-CFC.

## 2. Experimental

### 2.1. SO-CFC fabrication

An anode supported tubular SO-CFC was used for the long-term degradation test. To fabricate the SO-CFC, first an extruded porous tubular tube that served as the anode support for the tubular SO-CFC was manufactured. The other cell components were fabricated in thin layers and attached to 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, the well-dispersed paste was extruded in the form of a tubular anode support [22]. 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 the coating of the electrolyte film. The vacuum-slurry dip-coating method was used to coat a YSZ electrolyte layer onto the surface of the pre-sintered tubular anode support, which was then co-fired at 1400 °C. The cathode materials (La<sub>0.85</sub>Sr<sub>0.15</sub>)<sub>0.9</sub>MnO<sub>3</sub> (LSM) and La<sub>0.6</sub>Sr<sub>0.4</sub>Co<sub>0.2</sub>Fe<sub>0.8</sub>O<sub>3</sub> (LSCF) were synthesized by a solid-state powder reaction, for which the materials were weighed in the required proportions and mixed in ethanol by ball milling. The multilayered cathode, composed of an LSM/YSZ composite, LSM, and LSCF, was coated onto the surface of the YSZ electrolyte by slurry dip-coating method and co-sintered at 1150 °C. The active area of the single SO-CFC was 10 cm<sup>2</sup>. Details of the components and the microstructure of the tubular SO-CFC are shown in Fig. 1(a) and (b), respectively.

### 2.2. Characterization of the carbon fuel

To investigate the electrochemical characteristics and long-term degradation, activated carbon (Kuraray, Japan) was used as the carbon fuel. The coconut shell based powdered activated carbon fuel average particle size was ~100 mesh. The powdered activated carbon was used directly in the thermal analysis without any further processing. Proximate analysis of the fuel was performed using thermogravimetry (TGA-701, LECO Co., USA), and an elemental analyzer (TruSpec, LECO Co., USA) was employed for the ultimate analysis. The results of the proximate and ultimate analyses of the fuels are presented in Table 1. For the TGA/DTG analysis of the carbon fuel, SDT 2960 Simultaneous DTA-TGA (TA Instruments, USA) was used; this device had a measurement accuracy of ±2%. The heating rate of the samples during the TGA analysis was 10 °C/min and the atmosphere gas (air/Ar) flow rate was kept at 50 ml/min.

### 2.3. SO-CFC long-term degradation test

The schematics and an actual image of the experimental setup for the SO-CFC long-term testing are shown in Fig. 1 (c) and (d), respectively. A steam gasifier with a diameter of 6 cm and a height of 15 cm, with a provision of carbon refilling, was connected directly to the SO-CFC in the furnace. A micro pump (Eldex, Optos Pump model 2HM) was used to feed a controlled amount of water to a steam generator, which supplied steam to the bottom of the gasifier. At the exhaust of the gasifier, to determine the gas composition, one line was fed to an online gas chromatograph (PerkinElmer Clarus 580GC). Before the start of the long-term degradation test, the composition of the gasified fuel gas at the exit of the gasifier was monitored continuously for 40 h at 100 cc/min flow of steam and 750 °C. During this test, to observe any changes in the gas composition, carbon fuel was also refilled in the

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