



# Using potassium catalytic gasification to improve the performance of solid oxide direct carbon fuel cells: Experimental characterization and elementary reaction modeling



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## HIGHLIGHTS

- Potassium catalytic gasification based direct carbon fuel cell.
- Elementary reaction model and comprehensive experimental characterization.
- Effects of carbon bed geometries and operating parameters.

## ARTICLE INFO

### Article history:

Received 29 August 2013

Received in revised form

5 December 2013

Accepted 5 December 2013

Available online 14 December 2013

### Keywords:

Catalytic gasification

Direct carbon fuel cell

Elementary reaction

Modeling

Heterogeneous chemistry

## ABSTRACT

The performance of a solid oxide electrolyte direct carbon fuel cell (SO-DCFC) is limited by the slow carbon gasification kinetics at the typical operating temperatures of cell: 650–850 °C. To overcome such limitation, potassium salt is used as a catalyst to speed up the dry carbon gasification reactions, increasing the power density by five-fold at 700–850 °C. The cell performance is shown to be sensitive to the bed temperature, emphasizing the role of gasification rates and that of CO production. Given the finite bed size, the cell performance is time-dependent as the amount of CO available changes. A reduced elementary reaction mechanism for potassium-catalyzed carbon gasification was proposed using kinetic data obtained from the experimental measurements. A comprehensive model including the catalytic gasification reactions and CO electrochemistry is used to examine the impact of the catalytic carbon gasification process on the device performance. The power density is maximum around 50% of the OCV, where carbon utilization is also near maximum. Results show that bed height and porosity impact the power density; a thicker bed maintains the power almost constant for longer times while lower porosity delivers higher power density in the early stages.

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## 1. Introduction

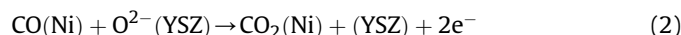
Fuel cells, especially those operating directly on fossil fuels, constitute a promising technology for electricity generation because of their high efficiency and lower emission. Direct carbon fuel cells (DCFCs) have the added advantage of consuming an abundant and relatively cheap fuel that can be derived directly from coal, as well as their potential for enabling the separation of CO<sub>2</sub> from the products without extra equipment or energy penalty. DCFCs are classified according to the electrolyte, i.e., molten

hydroxide [1–6], molten carbonate [7–11] or solid oxide electrolyte [12–19], although combining molten carbonate and solid oxide electrolyte has also been suggested [20–22]. In this paper, we focus on solid oxide electrolyte DCFC (SO-DCFC). SOFCs operate at high temperature, improving the electrochemical reactivity as well as the ion diffusivity through the electrolyte, and do not suffer from liquid electrolyte consumption and corrosion. On the other hand, operating at high temperature can increase the thermal stresses and possible material degradation.

The anode reaction mechanism of the SO-DCFC is more complicated than in the gas-fueled counterpart. This mechanism has been investigated in several studies. Nakagawa and Ishida [12] placed charcoal in the fuel chamber of a solid oxide fuel cell, 5 mm

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away from the anode and used nitrogen as the anode gas carrier. They ran the cells at 1075, 1180 and 1275 K. Using the experimental results, they concluded that the electrochemical reactions were driven by the carbon monoxide produced via charcoal gasification according to the following reactions:



**Reaction (1)**, the Boudouard reaction, is the carbon gasification reaction responsible for production of CO, while **Reaction (2)** is the electrochemical oxidation reaction of CO, which takes place at the anode triple phase boundary (TPB). **Reaction (3)** is the electrochemical reduction reaction at the cathode. Gür and Huggins [13] proposed an oxidation mechanism using an experiment in which they placed solid carbon adjacent to an yttria-stabilized-zirconia (YSZ) tube with platinum coated electrodes, and introduced helium as the anode gas. The temperatures of cell and carbon fuel were controlled independently.

In the experiments mentioned above, carbon can hardly be electrochemically oxidized directly (as in  $\text{C} + 2\text{O}^{2-} \rightarrow \text{CO}_2 + 4\text{e}^-$  or  $\text{C} + \text{O}^{2-} \rightarrow \text{CO} + 2\text{e}^-$ ), and therefore the anode reactions that determine the cell performance are **reactions (1) and (2)**, together. In the absence of an external gasification medium such as  $\text{CO}_2$ , the products of the electrochemical reaction act as a gasification medium. Moreover, in a typical SO-DCFC, the temperature of the carbon fuel is the same as that of the cell, ranging from 600 to 1000 °C [14]. As reported in our previous work [23], **reactions (1) and (2)** strongly depend on temperature. The carbon gasification reaction is slow below 800 °C and hence it is likely to be the rate-limiting step determining the overall performance of DCFC.

It has been widely demonstrated that alkali metals are effective catalysts for carbon gasification [24–28]. Those metals have been used in coal gasification to raise its rate at lower temperatures. In a series of experiments, Lee and Kim [29] measured the catalytic activities of alkali and transition metal salts. Moulijn and Kapteijn [30] proposed a possible structure of the active intermediates for the K-catalysis and demonstrated the catalytic process pathways. Using molecular dynamics calculations, Chen and Yang [31] developed a unified mechanism for dry and wet carbon gasification by  $\text{CO}_2$  and  $\text{H}_2\text{O}$ , respectively, using alkali metals as catalysts. They reported two kinds of oxygen intermediates/complexes dispersed in the alkali metal clusters, which could explain the observed phenomena that a catalyst changes the gasification rates without changing the activation energy. Struis et al. [32] and Huang et al. [33] suggested simplified models for catalytic carbon gasification processes and analyzing the kinetic behavior of metal catalyzed gasification.

There has been less effort to develop elementary reaction mechanisms for modeling carbon catalytic gasification, which is essential to better understand its role in improving the DCFC performance. In this paper, potassium was used to accelerate carbon gasification. A series of experiments were conducted using potassium salt embedded in the carbon fuel, while fixing the temperature of the fuel cell to 750 °C and varying that of carbon fuel from 700 to 850 °C, in order to determine the impact of catalytic gasification on the DCFC performance. A comprehensive elementary reaction mechanism of carbon catalytic gasification was introduced based in part on the work in Refs. [31,34,39–41]. Kinetic data for the model were obtained by fitting the model to the experimental results. Combined with the SO-DCFC model proposed by our group [36,37], the performance of the cell was simulated to gain insight

into the relative roles of gasification and electrochemistry, and to determine the impact of bed geometric design on the power density and carbon utilization.

The study reported in this paper utilizes a finite size carbon bed as the fuel, and examines the impact of the bed size on the power density and other cell performance characteristics as a function of time. As will be shown, given that the electrochemically active component here is CO, which is produced by the dry gasification of the carbon bed, and since the gasification rate depends on the bed geometry, the cell performance is also time-dependent. Moreover, conditions under which a steady performance can be achieved for a finite period of time are also explored.

In Section 2, we describe the experimental apparatus used to collect gasification kinetic and cell performance data. In Section 3, models for the gasification kinetics and the fuel cell performance are briefly described. Section 4 shows experimental and simulation results, as well as discussions regarding factors controlling cell performance. Conclusions are summarized in Section 5.

## 2. Experiment

### 2.1. Experimental setup

An anode-supported SOFC button cell fabricated by SICCAS (Shanghai Institute of Ceramics, Chinese Academy of Sciences) was used in this study. It consisted of a nickel/yttria-stabilized zirconia (Ni/YSZ) anode support layer (680 μm), a nickel/scandia-stabilized zirconia (Ni/ScSZ) anode active interlayer (15 μm), a ScSZ electrolyte layer (20 μm), and a lanthanum strontium manganite (LSM)/ScSZ cathode layer (15 μm). The diameter of the cathode layer was 1.3 cm and that of other layers was 2.6 cm.

To examine the effect of catalytic gasification on the SO-DCFC performance, the carbon fuel and the button cell were separated to avoid possible carbon direct electrochemical oxidization. Fig. 1(a) and (b) shows schematics of the test equipment. Located at the end of two coaxial alumina tubes, the button cell was supported by a horizontal alumina plate which was constrained by springs. The alumina plate with a hole in the middle on the anode side offered a channel for the anode fuel gases. A platinum (Pt) mesh was used as cathode current collector. The oxidant flowed into the inner tube to the cathode and passed through the porous Pt mesh. A Ni felt (thickness 2 mm) was fixed to the anode support layer with silver paste to collect the anodic current. The carrier/gasification gas was introduced into the carbon bed, which was contained in another alumina tube. For both the anode and cathode, Pt wires were used as voltage and current probes. The horizontal alumina plate had a through-hole of the same diameter as the cell cathode, and it was used to constrain the carbon bed. The layout of the carbon bed is exhibited in Fig. 1(a). The carbon fuel was placed in another small quartz tube under the button cell. A porous plate sintered of quartz sand was fixed to the quartz tube. The carbon fuel, quartz wool and corundum ceramic chips were placed on the plate sequentially. The anode gas flowed into the small quartz tube, through the porous plate, carbon fuel, quartz wool, alumina chips and finally to the button cell anode. The quartz wool and corundum ceramic chips were used to prevent the carbon fuel powder from being blown away by anode gas.

The device was enclosed in a quartz tube and heated by a furnace to the required temperature. Pure  $\text{H}_2$  passed through the chamber for 1 h to fully reduce the anode at a flow rate of  $50 \text{ scc min}^{-1}$  (Standard Cubic Centimeter per Minute;  $8.33 \times 10^{-7} \text{ m}^3 \text{ s}^{-1}$ ).

### 2.2. Catalyst addition and fuel preparation

Commercial carbon black (Black Pearls 2000, GP-3848, Cabot Corporation, Boston, MA), with 94.61% pure carbon, was used as a

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