



## Origin of peculiar electrochemical phenomena in direct carbon fuel cells

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## HIGHLIGHTS

- The physicochemical properties of a carbon dictates its gasification tendency.
- CO is produced most predominantly due to the reverse Boudouard reaction.
- DCFC operates effectively if the number of CO is balanced with that of O<sup>2-</sup>.
- Methodologies to overcome or alleviate peculiarities are suggested.

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## ABSTRACT

Solid oxide carbon fuel cells (SO-CFCs), which are a part of direct carbon fuel cells (DCFCs), adopt the same chemistry of solid oxide fuel cells (SOFCs). Despite their same chemistry, a difference in electrochemical behaviors has been observed. Unlike SOFCs where gas is imposed as fuel, SO-CFCs employ solid-state carbonaceous fuel that undergoes gasification and produces gaseous fuel such as carbon monoxide (CO). The amount of CO production varies according to the physicochemical properties of carbon source. In this contribution, the authors prove that the plethora or lack of CO in SO-CFCs can cause poisoning on Ni electrocatalyst and thereby a decrease in electrochemical performance. The authors demonstrate poisoning mechanisms and they finally suggest methodologies to overcome or alleviate the poisonings.

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## 1. Introduction

Our generation struggles to constitute milestones on the road towards an environmentally sustainable society. As part of efforts to build the sustainable society, scientists intensify researches on renewable energy sources [1]. Fuel cells are a part of the recent movement to replace conventional power generation systems. Fuel cells come in many varieties; however, they all share the same advantages—they are not subject to the Carnot limitations, and they undergo fewer intermediate conversion steps compared with the conventional systems [2–4]. Among the varieties, direct carbon fuel cells (DCFCs), in particular solid oxide carbon fuel cells (SO-CFCs), are a suggestion for deploying solid carbonaceous materials [5–7], thanks to such benefits: high efficiency, structural simplicity, and fuel flexibility [8–21].

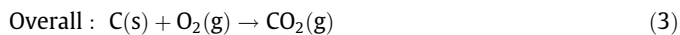
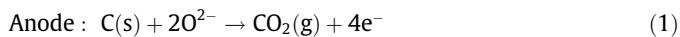
The terminology “SO-CFC” was coined to describe a fuel cell system that involves “solid oxide materials or ceramics” as electrolyte, and “solid carbonaceous materials” as fuel. [6] In general, carbon fuel is located on an anode surface, and it is later oxidized in order to produce electricity. O<sup>2-</sup> ions, which evolved from O<sub>2</sub> gases at a cathode, are the oxidant and then they travel to the anode surface through an electrolyte. In other words, the same system holds for both SO-CFCs and solid oxide fuel cells (SOFCs) except that the latter produces electricity directly from oxidizing gas-phase fuel [22,23]. By the same token, SO-CFCs have been widely believed that solid carbon is directly oxidized as fuel (Eqs. (1)–(3)). However, it is recently identified that the solid carbon mostly serves as a precursor of actual fuel such that it undergoes gasification and produces further oxidizable gaseous molecules [24–30]. In particular, the reverse Boudouard reaction (Eq. (4)) contributes largely to the production of the actual fuel such as CO in SO-CFCs [29]. As compared with SOFCs, SO-CFCs generally have a more CO rich atmosphere [23,31,32]. In this regard, a new mechanism explaining SO-CFCs is proposed as described in Eqs.

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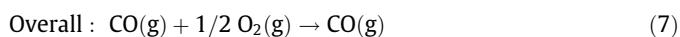
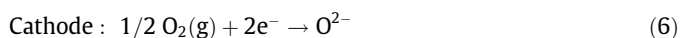
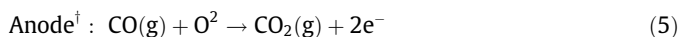
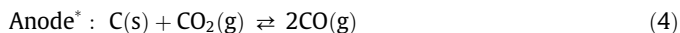
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(4)–(7). Based on the equations, SO-CFCs have a two-step process at the anode surface, namely i) gasification reaction to produce actual fuel, and subsequently, ii) electrochemical reaction to generate electricity.

Traditional mechanism in SO-CFCs



Recently proposed mechanism in SO-CFCs



\*The first (preceding) step. Occurring within an anode chamber. The reverse Boudouard reaction ( $K_{\text{eq}} = 0$  at 702 °C). Eq. (4) can be replaced by any reaction producing further oxidizable gaseous molecules.

†The following step. Occurring at electrochemical reaction sites (triple phase boundaries). Eq. (5) can be modified according to Eq. (4), e.g. Eq. (5) should be modified as a six-electron transfer reaction if Eq. (4) is replaced by methanation.

Researchers on SO-CFC may encounter various peculiarities that differ from SOFCs and which can hinder them from correctly measuring fuel cell performances. The cause of these peculiarities is still not identified. We hypothesized that the discrepancy between SO-CFCs and SOFCs is attributed to a common misunderstanding about the anode reactions in SO-CFCs (Eq. (1) vs. Eqs. (4) and (5)—solid carbon reacts directly with  $\text{O}^{2-}$  ions. In this regard, we assumed that the peculiarities stemmed from the two-step behav-

ior at carbon/anode interfaces, in particular, from an imbalance between the first and the following steps (e.g. Eqs. (4) and (5)). One can impose the imbalance by either controlling the first step or the following one, thereby demonstrating whether the assumption holds true. As regards the first reaction, the imbalance is obtainable by changing a carbon source because the physicochemical properties of carbon could affect gasification reactivity. As for the following reaction, the imbalance can take place by manipulating the rate of  $\text{O}_2$  supply towards a cathode, which in turn will result in the fewer or greater number of  $\text{O}^{2-}$  ions arriving at an anode surface. In this contribution, we investigated how the two imbalances affect the electrochemical performance of SO-CFC, and how to control or overcome the imbalances.

## 2. Experimental

### 2.1. Materials and methods

#### 2.1.1. Cell features

Single cells, consisting of four layers stacked together, were purchased from KCeracell. The cells have a button-type feature and they were anode-supported. The detailed layer profile is described in Table 1. Ni is the most commonly used electrocatalyst in both SOFCs and SO-CFCs because of its high stability at elevated temperatures, excellent electric conductivity, relatively lower price than precious metals, and less vulnerability to CO poisoning than Pt which is a typical electrocatalyst in low-temperature fuel cells [33–36]. Even though as-purchased cells contain NiO in place of a Ni electrocatalyst, an anode layer can be reduced into Ni with the help of fuel additives and/or oxidizable gaseous molecules at elevated temperatures. When works were undertaken using our experimental set-up and apparatus, it was assumed that the cells were converted to metallic Ni at ca. 550 °C [37,38] because we observed that a resistance value began to dramatically decrease by more than 2 orders of magnitude at that temperature region.

**Table 1**  
Detailed layer profile of a single cell employed in this work.

	Anode	Electrolyte	Interlayer	Cathode
Material	NiO-YSZ	YSZ	LSM-YSZ	LSM
Structure	Porous	Dense	Porous	Porous
Thickness	≈550 μm	≈14.5 μm	≈14.9 μm	≈16.7 μm

Note: YSZ stands for yttria-stabilized zirconia; LSM stands for lanthanum strontium manganite.

**Table 2**  
General information of carbon samples and their experimental conditions.

Sample	Code	Bulk density/g cm <sup>-3</sup>	Viscosity <sup>a</sup> /cps	Quantity <sup>b</sup> /g	Carbon content <sup>c</sup> /at.%
Black Pearl 2000	BP	0.149	125.8	0.6	98.65
Ketjen black EC-600JD	KB	0.12	500.5	0.25	97.93
Timcal 350G	T3	0.135	92.4	0.49	98.33
Vulcan XC-72	VX	0.295	48.0	1.1	98.58
Timcal 150G	T1	0.218	43.5	1.2	99.13
Acetylene black	AB	0.146	93.5	0.8	98.43
CNT M95	CNT	0.022	3075.6	0.2	95.67
Graphene Nanoplatelet C750	GNP	0.712	40.7	1.5	98.52
Graphite (Synthetic, <20 μm)	GR	0.242	32.1	3.4	98.84

<sup>a</sup> The viscosity value of the colloidal mixture which comprised of 400 mg of each carbon sample and 40 mL of ethylene glycol was measured at room temperature. As reference, the viscosity value of ethylene glycol was also measured, which was 28.6 cps.

<sup>b</sup> The required mass of each carbon sample to make it exhibit plasticity (being dough-like or clay-like) when 3 mL of ethylene glycol is added as a binder. In general, the order of the mass values is inversely proportional to that of the viscosity values.

<sup>c</sup> Extracted from EDX (Oxford, INCAx) data unless otherwise specified. In the case of CNT, the value is extracted from another EDX device (EMAX 7200-H, Horiba). Note: Aside from CNT, carbon samples contains very low sulfur contents (the sulfur contents of all of these samples are insignificant) CNT resulted in considerable ash contents, and its ash also underwent EDX, giving the following results: O (54.89 at.%), Al (15.89 at.%), Si (4.55 at.%), S (4.89 at.%) and Fe (19.78 at.%) Sulfur could result in a poisoning to a cell when exposed for a long term to the cell [39].

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