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Probing novel triple phase conducting composite cathode for high performance protonic ceramic fuel cells

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ABSTRACT

A key obstacle to practical operation of protonic ceramic fuel cells (PCFCs) is the development of high efficient cathode materials. In this study, we report the identification of $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{2-\delta}$ (GDC) infiltrated $\text{PrBaCo}_2\text{O}_{5+\delta}$ (PBC) – $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2}\text{O}_{3-\delta}$ (BZCY) materials as a novel triple phase ($\text{e}^-/\text{H}^+/\text{O}^{2-}$) conducting composite cathode of PCFCs. This triple phase conducting composite cathode is confirmed to promise the extended electrochemical active sites and the accelerated oxygen reduction process. The polarization resistance (such as $0.051 \Omega \text{ cm}^2$ at 700°C under open circuit voltage) of GDC infiltrated PBC-BZCY electrode is lower than other reported PCFCs, indicating the advanced electrochemical performance. The encouraging result is definitely a significant progress for the PCFCs field, which demonstrates that desirable electrode performance can be realized by applying triple phase conducting composite electrode.

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Introduction

Ceramic fuel cells (CFCs) possess several attractive advantages such as high energy conversion efficiency, low pollutant emission, good fuel flexibility, and efficient reclamation of waste heat [1–4]. However, high operating temperature of CFCs ($800\text{--}1000^\circ\text{C}$) is an obstacle to commercialization, mainly due to significant degradation, sealing issue, as well as high thermal stress. In recent years, extensive efforts [5–10]

have been devoted to lowering the operating temperature of CFCs to intermediate temperature ($600\text{--}800^\circ\text{C}$), in order to promise flexibility of materials choice and avably reduce the cost of CFCs system. Compared with intermediate temperature CFCs (IT-CFCs) based on oxygen ion conducting electrolytes, such as doped ceria [11,12], CFCs using proton conducting electrolytes in general can offer quite a few advantages, including high protonic conductivity, low activation energy of protonic transport, good fuel utilization and high Nernst potential [13–16]. In the search of applicable protonic

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ceramic electrolytes, zirconium-stabilized barium cerates, typically $\text{Ba}(\text{Zr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2})\text{O}_{3-\delta}$ (BZCY) [17,18] and $\text{Ba}(\text{Zr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2-x}\text{Yb}_x)\text{O}_{3-\delta}$ (BZCYb) perovskites [3], have been reported to build outstanding balances between protonic conductivity and chemical stability over a wide operating conditions of IT-CFCs. The performances of these protonic ceramic fuel cells (PCFCs), however, are still inadequate for practical application primarily due to the lack of highly efficient cathode materials [14,19]. Therefore, it is vital to further develop PCFCs considering highly active and stable cathode materials.

In typical PCFCs system, hydrogen in the anode area is oxidized to create mobile protons in the form of a covalent bond with lattice oxygen atoms. The mobile protons transfer through the dense electrolyte into the cathode and undergo a half-cell reaction with oxygen to produce water. The overall half reaction of anode and cathode is expressed as Eqs. (1) and (2), respectively [14,20–22],



Where H_2 , O_2 , and H_2O represent hydrogen, oxygen gas and water vapor, O_o^\times is oxygen in lattice of electrolyte materials, $\text{OH}_\text{o}^\times$ stands for proton attached to the lattice oxygen of electrolyte, and e^- is electron. The primary function of the cathode is to facilitate oxygen dissociated adsorption process and to accelerate the migration of formed oxygen species. Mixed oxygen ionic and electronic conductors (MIECs), including $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (LSCF) [23], $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{O}_{3-\delta}$ (SSC) [24], $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (BSCF) [6], and $\text{PrBaCo}_2\text{O}_{5+\delta}$ (PBC) [25], are highly recommended as the cathode materials because of their excellent catalytic activities for the oxygen reduction reaction (ORR). However, Eq. (2) indicates that the water is produced in the vicinity of the three phase boundaries (TPB), where proton ($\text{OH}_\text{o}^\times$), electron, and oxygen molecular are available. Therefore, protonic conduction among the cathode is vital to enhance the cathode reaction of PCFCs due to the extended TPB from the interface of electrolyte/cathode to the cathode bulk. For instance, introducing proton conductor of $\text{BaCe}_{0.8}\text{Sm}_{0.2}\text{O}_{3-\delta}$ (BCS) into $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$ (SSC) electro-catalyst could effectively reduce the polarization resistance (R_p) of single cell from $3.10 \Omega \text{ cm}^2$ to $0.67 \Omega \text{ cm}^2$ at 600°C [26]. The similar effects were also identified by $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (LSCF) – $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2}\text{O}_{3-\delta}$ (BZCY) [27] and LSCF – $\text{BaCe}_{0.9}\text{Y}_{0.1}\text{O}_3$ [28] composite cathodes. Furthermore, with the development of the fundamental study on cathodes for PCFCs, several suggested elementary reaction steps of PCFCs cathode are proposed (Table S1) [29–31]. These models indicate that not only protonic conduction but also oxygen species transfer plays the positive role for the cathode reaction dynamics. Thus, new approaches to the tailoring of cathode materials for PCFCs should be considered.

In this regard, high conduction of electrons, protons, and oxygenic ions becomes one of the basic requirements for cathode materials of PCFCs. For example, Kim et al. [31] proposed a triple phase ($\text{e}^-/\text{H}^+/\text{O}^{2-}$) conducting layered perovskite

$\text{NdBa}_{0.5}\text{Sr}_{0.5}\text{Co}_{1.5}\text{Fe}_{0.5}\text{O}_{3+\delta}$ (NBSCF) as a single phase cathode of PCFCs, which presented a superior electrochemical performance. Duan et al. [10] developed a triple phase conducting $\text{BaCo}_{0.4}\text{Fe}_{0.4}\text{Zr}_{0.1}\text{Y}_{0.1}\text{O}_{3-\delta}$ (BCFZY0.1) cathode, which greatly improved oxygen reduction reaction kinetics at intermediate to low temperatures. Considering quite limited materials selection for developing single phase cathodes with triple phase conducting, the strategy of designing triple phase conducting composite materials come into another story for highly efficient cathode materials of PCFCs.

In this study, we reported on the application of a novel triple phase conducting composite cathodes, $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{2-\delta}$ (GDC) infiltrated $\text{PrBaCo}_2\text{O}_{5+\delta}$ (PBC)- $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2}\text{O}_{3-\delta}$ (BZCY), for PCFCs. The triple phase conducting cathode was demonstrated to largely extend the electrochemical active sites and effectively accelerate oxygen reduction process, as proposed in Fig. 1. Using 29.6 wt.% GDC infiltrated PBCO-BZCY cathode, single cell based on BZCY electrolyte provided a superior electrochemical performance with a peak power density of 1.02 W cm^{-2} and a low polarization resistance of $0.051 \Omega \text{ cm}^2$ at 700°C . Moreover, no obvious degradation of the cell performance was observed within 100 h testing.

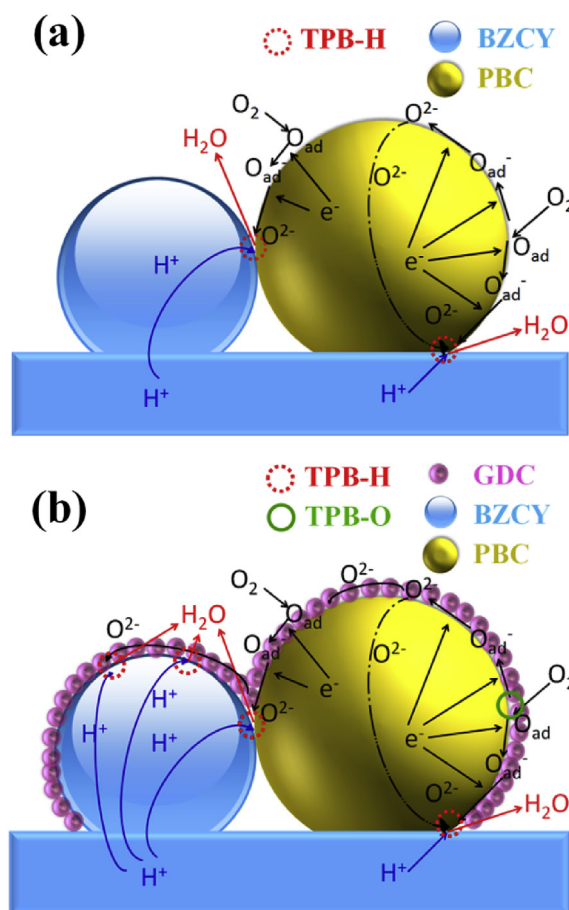


Fig. 1 – Schematic illustration of the cathode reaction of PCFCs using (a) PBC-BZCY and (b) GDC infiltrated PBC-BZCY.

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