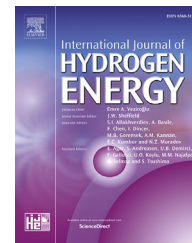




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## Short Communication

# Stable $\text{SrCo}_{0.7}\text{Fe}_{0.2}\text{Zr}_{0.1}\text{O}_{3-\delta}$ cathode material for proton conducting solid oxide fuel cell reactors

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## ABSTRACT

$\text{SrCo}_{0.7}\text{Fe}_{0.2}\text{Zr}_{0.1}\text{O}_{3-\delta}$  (SCFZ) perovskite is prepared using a combustion method. SCFZ exhibits high stability while  $\text{SrCo}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$  without Zr doping decomposes in  $\text{CO}_2$ - and  $\text{H}_2\text{O}$ -containing atmosphere at elevated temperature. SCFZ also displays excellent chemical compatibility with  $\text{BaCe}_{0.7}\text{Y}_{0.2}\text{Zr}_{0.1}\text{O}_{3-\delta}$  (BCYZ) proton conductor. A ceramic membrane fuel cell reactor is assembled with SCFZ + BCYZ composite cathode, porous Pt anode and BCYZ electrolyte. High selective ethylene and electrical energy are co-generated from ethane in the proton conducting solid oxide fuel cell reactor.

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## Introduction

Proton-conducting solid oxide fuel cells (SOFCs) have attracted increasing attentions due to their higher ionic conductivity than oxygen ion conductors at low or intermediate temperature [1–3], higher energy efficiency [4–6] and hydrocarbons can be simultaneously converted to valuable alkenes and electricity while without  $\text{CO}_2$  greenhouse gas emission [7–11]. Ethylene as the simplest alkene is the most produced organic compound in the world which is typically manufactured by steam cracking of hydrocarbons at present. However,

this dehydrogenation process consumes large amounts of energy. In contrast, oxidative dehydrogenation of ethane to ethylene is an exothermic reaction. But ethane is oxidized readily and completely to  $\text{CO}_2$  when ethane and oxygen are contacted and so it is difficult to achieve high selectivity to ethylene [7]. Proton conducting SOFC reactors with Pt electrodes have been developed for electrochemically oxidative dehydrogenation of ethane to high selective ethylene and co-generation of electricity [9,12]. Cathode for oxygen reduction plays an important role in the performance of SOFCs. Conventional SOFC cathode of  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-\delta}$  has low catalytic activity and electrical conductivity at temperature lower than

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800 °C thus it is not suitable to use in these SOFCs [13–15]. Cobaltite oxides such as  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$  [16],  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{1-x}\text{Fe}_x\text{O}_{3-\delta}$  [17],  $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$  [18],  $\text{PrBaCo}_2\text{O}_{5+\delta}$  [19] have high performance as cathode catalyst in intermediate or low temperature SOFCs. Unfortunately, these oxides are not stable in  $\text{CO}_2$  and humid atmospheres which is a drawback for their practical applications in SOFCs [20–22] especially in proton conducting SOFCs which water is formed at the cathode. More recently, Nb doping for the B-site greatly improved the chemical stability and  $\text{CO}_2$  tolerance of  $\text{SrCoO}_{3-\delta}$  materials [23]. There are very few reports describing stable cathode materials for proton conducting SOFCs although a lot of high active cathode materials have been developed to use in SOFCs based on proton conductors recently [2,24–27].

Here, we report a stable  $\text{SrCo}_{0.7}\text{Fe}_{0.2}\text{Zr}_{0.1}\text{O}_{3-\delta}$  (SCFZ) cathode material for proton conducting SOFCs, and in particular for use in hydrocarbon SOFC reactors to co-generate electrical power and ethylene from ethane.

## Experimental

SCFZ powders were synthesized through a combustion method.  $\text{Sr}(\text{NO}_3)_2$ ,  $\text{Co}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Fe}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  and  $\text{ZrO}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  were stoichiometrically weighed, dissolved in deionized water and heated at 90 °C in a glass beaker. Then citric acid was added with a molar ratio of 2:1 total metal ion. Until the resulting solution was becoming a dry gel. The dry gel was combusted at 400 °C and then calcined at 1000 °C determined through thermal gravity analysis using a TA SDT Q600 instrument.

BCYZ powders were prepared by a solid state reaction method.  $\text{BaCO}_3$ ,  $\text{CeO}_2$ ,  $\text{Y}_2\text{O}_3$  and  $\text{ZrO}_2$  powders were stoichiometrically weighed and mixed by ball-milling for 24 h, followed by calcination at 1300 °C for 10 h in air. The resulting materials were pressed at 5 tons into discs with 1 mm thickness, and sintered at 1500 °C for 10 h in air.

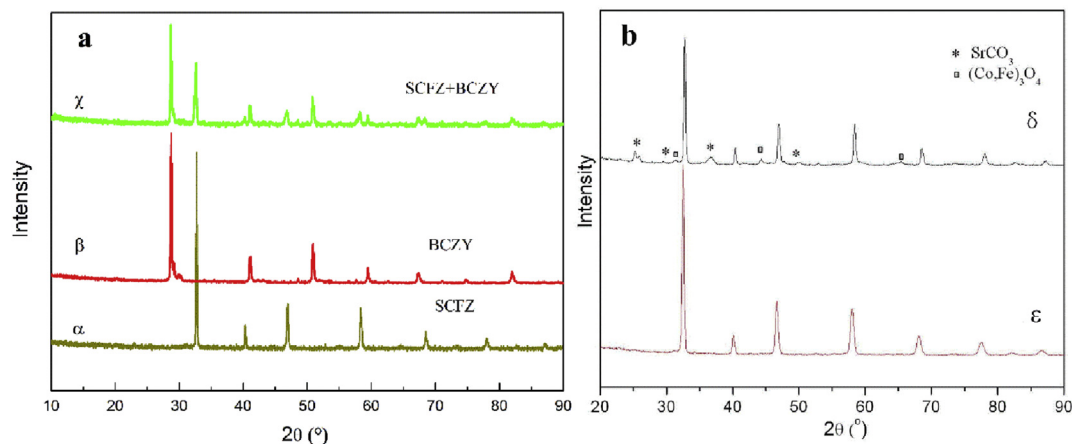
An intimate mixture of 50 wt% SCFZ and 50 wt% BCYZ powders was screen printed onto the surface of the electrolyte and sintered at 1000 °C for 5 h to form a 0.5  $\text{cm}^2$  cathode. Pt

paste was applied onto the opposite side of the electrolyte, then heated at 900 °C for 30 min to bond the components and so obtain the membrane electrode assembly (MEA). Au paste and mesh were used as current collector at the anode and cathode. Ethane was fed into the anode compartment and air was the cathode feed.

The phase structures of materials were identified using a Rigaku Rotaflex X-ray diffractometer (XRD) with  $\text{Co K}\alpha$  radiation. Morphologies of samples were determined using a Hitachi S-2700 scanning electron microscope (SEM). Cell performance tests were measured using a Solartron 1287 electrochemical interface together with 1255B frequency response analysis instrumentation.

## Results and discussion

Fig. 1a shows the XRD patterns for the as-prepared SCFZ and BCZY powders and their mixtures after chemical compatibility test. The SCFZ and BCZY oxide crystallize in a cubic  $\text{ABO}_3$  perovskite-type structure. After the SCFZ and BCZY mixture are calcined at 1100 °C for 10 h, the SCFZ and BCZY samples still retain their own structures without any secondary phase and peak shift. This suggests a good chemical compatibility between SCFZ and BCZY when the calcining temperature is below 1100 °C. Fig. 1b displays that both SCFZ and  $\text{SrCo}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$  (SCF) powders are pure perovskite oxides with similar structures, suggesting that Zr is intimately doped into the parent structure of SCF. There is a small concentration of  $\text{CO}_2$  in the cathode air feed, and water is formed in the cathode compartment when protons transported through electrolyte to reduce the oxygen. Thus the cathode material for the proton conducting SOFC reactors must be stable in  $\text{CO}_2$ - and  $\text{H}_2\text{O}$ - containing atmospheres. Fig. 1b indicates that SCF decomposes to  $(\text{Co,Fe})_3\text{O}_4$  and  $\text{SrCO}_3$  while SCFZ retains its perovskite structure after treatment in  $\text{CO}_2$  and  $\text{H}_2\text{O}$  containing oxygen at 700 °C for 10 h. The results suggest that Zr doping could enhance the stability of the cobalt-based perovskite oxide, which is similar to the improvement of chemical stability of  $\text{BaCo}_{0.4}\text{Fe}_{0.6-x}\text{Zr}_x\text{O}_{3-\delta}$  perovskite [28].



**Fig. 1** – (a) XRD patterns of (α) as-prepared SCFZ (β) as-prepared BCYZ and (γ) the mixture of SCFZ and BCYZ after sintering at 1100 °C for 10 h; (b) XRD patterns of (δ) SCF and (ε) SCFZ cathode materials after exposure to 5%  $\text{CO}_2$  + 3%  $\text{H}_2\text{O}$  (balance with  $\text{O}_2$ ) at 700 °C for 10 h.

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