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Short Communication

A novel bifunctional catalyst of $\text{Ba}_{0.9}\text{Co}_{0.5}\text{Fe}_{0.4}\text{Nb}_{0.1}\text{O}_{3-\delta}$ perovskite for lithium–air battery

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ABSTRACT

$\text{Ba}_{0.9}\text{Co}_{0.5}\text{Fe}_{0.4}\text{Nb}_{0.1}\text{O}_3$ (BCFN) perovskite has been synthesized by a solid-state reaction method, and characterized by XRD, SEM, BET. This oxide has a porous structure and a specific surface area of $10.24 \text{ m}^2 \text{ g}^{-1}$ after ball-milled 24 h. The catalytic activity of the oxide for the oxygen reduction reaction (ORR) and the oxygen evolution reaction (OER) in 0.1 M KOH solution has been studied by using a rotating ring-disk electrode (RRDE) technique. RRDE results show that the ORR mainly favors a direct four electron pathway, and a maximum cathodic current density of -5.70 mA cm^{-2} at 2500 rpm was obtained, which is close to that of Pt/C (20 wt.% Pt on carbon) electrocatalyst in the same testing conditions. Compared with behaviors of pure C and Pt/C electrode, a lower onset potential of BCFN for OER is observed, and a bigger anodic current at the same applied potential is obtained. Considering small surface area of the BCFN catalyst, a big overpotential is given in the discharge–charge curves. However, the outputs of 2032 coin Li–air batteries in a dry gas mixture composed of 80 vol.% pure N_2 and 20 vol.% pure O_2 demonstrated that BCFN could be a potential bifunctional catalyst for the Li–air battery.

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1. Introduction

There is growing interest in hybrid electric vehicles requiring smaller and lighter weight batteries to meet today's energy and environmental challenges. Lithium–air battery is considered as the most attractive contender for such applications as it has the highest theoretical specific energy density

of 5200 Wh kg^{-1} (including oxygen) [1,2]. Li–air batteries are compact, lightweight and cost effective because they adopt cheap and light oxygen during discharge, replacing expensive chemical constituents used in current rechargeable batteries. For the lithium–air batteries, the air cathode is the most serious challenge for eventual development because of the sluggish kinetics of oxygen reduction reaction (ORR) during discharging process and oxygen evolution reaction (OER)

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during charging process. The sluggish kinetics of ORR and OER in lithium–air batteries are ascribed to the low efficiency of catalysts [3]. The performance of lithium–air batteries can be drastically improved by incorporating an efficient catalyst to achieve lower polarization [4]. Therefore, it is important to develop highly efficient catalysts for the ORR and the OER in air cathode to realize commercial lithium–air batteries.

Perovskite oxide exhibits good cation ordering, which can provide disorder-free channels of oxygen vacancies to enhance the mobility of oxygen ions [5–7]. As we all know that perovskite oxides have wide applications in catalysis for fuel cells and metal–air batteries due to their defective structures and excellent oxygen mobility. In recent years, some perovskite cathode materials of solid oxide fuel cell (SOFC) have successively been reported as bifunctional catalyst in Li–air batteries. Yang [8] prepared $\text{Sr}_{0.95}\text{Ce}_{0.05}\text{CoO}_3$ perovskite oxide loaded with copper nanoparticles and applied it as a bifunctional catalyst for Li–air battery with aqueous/organic mixed electrolyte. Zhao [9] prepared hierarchical mesoporous perovskite $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_{2.91}$ nanowires and used it as bifunctional catalyst in lithium–air batteries, which exhibited ultrahigh capacity over 11,000 mAh g^{-1} . As one of the most frequently-used cathode materials of solid oxide fuel cells, $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ perovskite oxide has also demonstrated promising catalytic properties in Li–air battery with non-aqueous electrolyte in many groups' works [10,11].

$\text{Ba}_{0.9}\text{Co}_{0.5}\text{Fe}_{0.4}\text{Nb}_{0.1}\text{O}_3$ (BCFN), which has been used as an efficient SOFC cathode material [12,13], might also possess bifunctional catalytic activities desirable for Li–air battery cathode application. In this communication, we attempt to assess the prospective of BCFN as bifunctional catalyst in the air cathode by investigating the electrochemical performances in terms of ORR and OER, as well as cyclic life in alkaline media through a rotating-ring-disk electrode (RRDE) testing. Furthermore, a 2032 coin Li–air battery was also assembled and tested with BCFN as cathode catalyst.

2. Experimental

$\text{Ba}_{0.9}\text{Co}_{0.5}\text{Fe}_{0.4}\text{Nb}_{0.1}\text{O}_3$ (BCFN) was prepared using a solid-state reaction method [12,13]. All starting materials of BaCO_3 , Co_3O_4 , Fe_2O_3 and Nb_2O_5 were of analytical grade and ordered from Guoyao Chemical Reagent Co. Ltd. The ball-milled mixture of the starting materials was calcined in air at 1000°C for 24 h.

The crystalline structure of BCFN was confirmed using a Philips X-ray diffractometer (Model PW1830). The morphology was examined in a scanning electron microscopy (FEI Quanta 200). The specific surface area of the oxide was determined by using a Brunauer–Emmett–Teller (BET) analysis system with an N_2 adsorptive medium.

The electrocatalytic activities for the ORR and OER were studied with a rotating ring-disk electrode (RRDE) technique using a Pine Electrochemical system (AFMSRX rotator, and AFCBP1 bipotentiostat) as that our previous described [7,14]. The RRDE electrode consisted of a catalyst film-coated GC disk (0.196 cm^2 of geometric surface area) surrounded by a Pt ring

(0.125 cm^2 of geometric surface area). Electrochemical experiments were carried out in a standard 3-electrode electrochemical cell at room temperature. A Pt-wire was used as the counter electrode, and an Ag/AgCl (1 M Cl^-) reference electrode was used in a double-junction reference chamber. The electrolyte was 0.1 M KOH solution prepared with ultrapure water (Millipore, $18.2\text{ M}\Omega\text{ cm}$). A BCFN oxide and acetylene black mixture was used as electrocatalyst for the ORR and the OER.

The catalyst ink of the dispersed BCFN oxide was made by mixing 10 mg oxide powder (50 wt.%) and 10 mg acetylene black with $190\text{ }\mu\text{l}$ Nafion solution (6% in ethanol, Aldrich) and $900\text{ }\mu\text{l}$ ethanol in an ultrasonic bath for 2 h. The working electrode was prepared by applying $7\text{ }\mu\text{l}$ catalyst ink onto the surface of GC electrode with a micropipette and drying in air for 0.5 h. The catalyst loading was 0.655 mg cm^{-2} .

Rotating-ring-disk-electrode (RRDE) voltammetric experiments were performed for ORR test with a 10 mV s^{-1} scan rate in O_2 saturated 0.1 M KOH solution at room temperature. The voltage scanning range was from 0.2 to -1.0 V vs. Ag/AgCl. The ring potential was set at 0.5 V vs. Ag/AgCl, which is considered to be sufficiently high to induce complete peroxide decomposition as reported elsewhere [15]. During the ORR test in O_2 saturated KOH solution, the background capacitive current contribution (obtained from N_2 saturated experiment) is subtracted from each voltammogram. For OER test, the voltage scanning range was from 0 to 1.0 V vs. Ag/AgCl. Prior to each

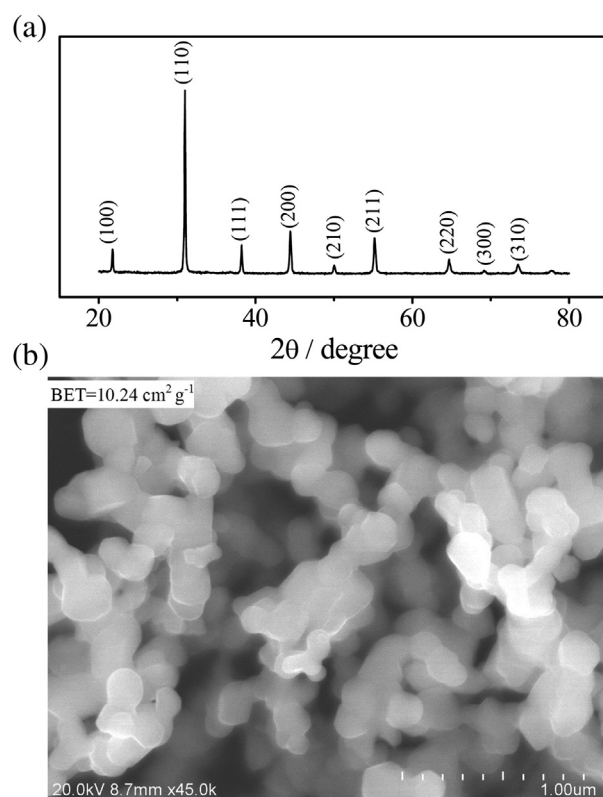


Fig. 1 – XRD pattern of BCFN powders (a); SEM image of BCFN powders (b).

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