



Strategy towards cost-effective low-temperature solid oxide fuel cells: A mixed-conductive membrane comprised of natural minerals and perovskite oxide



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HIGHLIGHTS

- Mixed-conductive membrane hematite/LCP-LSCF was applied to SOFC for the first time.
- Hematite/LCP-LSCF was prepared from natural minerals and perovskite via facile way.
- The ionic conductivity of the composite reached 0.153 S cm^{-1} at $600 \text{ }^\circ\text{C}$.
- SOFCs based on such composite exhibited excellent performances at low temperatures.

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ABSTRACT

Our previous work has revealed the feasibility of natural hematite as an electrolyte material for solid oxide fuel cells (SOFCs), tailoring SOFCs to be a more economically competitive energy conversion technology. In the present work, with the aim of exploring more practical uses of natural minerals, a novel composite hematite/LaCePrO_x-La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (hematite/LCP-LSCF) has been developed from natural hematite ore, rare-earth mineral LaCePr-carbonate, and perovskite oxide LSCF as a functional membrane in SOFCs. The heterogeneity, nanostructure and mixed-conductive property of the composite were investigated. The results showed that the hematite/LCP-30 wt% LSCF composite possessed balanced ionic and electronic conductivities, with an ionic conductivity as high as 0.153 S cm^{-1} at $600 \text{ }^\circ\text{C}$. The as-designed fuel cell using the hematite/LCP-LSCF membrane exhibited encouraging power outputs of $303 - 662 \text{ mW cm}^{-2}$ at $500 - 600 \text{ }^\circ\text{C}$. These findings show that the hematite/LCP-LSCF based fuel cell is a viable strategy for developing cost-effective and practical low-temperature SOFCs (LTSOFCs).

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1. Introduction

The demand for clean and renewable energy has stimulated vast efforts in applied energy conversion technologies. As a promising candidate, solid oxide fuel cell (SOFC) technology has attracted tremendous attention owing to its superiorities of high energy

conversion efficiency, fuel flexibility, and low pollution emissions [1–4]. Typically, SOFCs based on yttria-stabilized zirconia (YSZ) require high operating temperatures of over $650 \text{ }^\circ\text{C}$ to reach a sufficient ionic conductivity in the YSZ electrolyte [5,6]. In this regard, it is crucial to explore advanced alternative materials with sufficiently high ionic conductivity at reduced temperatures ($300 - 600 \text{ }^\circ\text{C}$) [7].

Natural minerals are abundant resources on earth that have been applied to energy conversion technologies [8–10]. The utilization of natural minerals as functional materials has enabled the direct conversion from thermal and chemical energies into electrical energy [11–13]. For instance, the natural minerals

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tetrahedrite ($\text{Cu}_{12-x}\text{M}_x\text{Sb}_4\text{S}_{13}$) and tennantite ($\text{Cu}_{12-x}\text{M}_x\text{As}_4\text{S}_{13}$) have been utilized as a direct source of thermoelectric materials for power generation [14]. An industrial-grade rare-earth mineral, LaCePr-carbonate (LCP-carbonate) has shown enormous potential to produce LaCePr-oxides (LCP-oxides) for electrolyte uses in SOFCs [15]. Among these studies on minerals, a natural hematite ($\alpha\text{-Fe}_2\text{O}_3$) mineral has emerged recently, showing great potentials in the field of solid state ionics. The natural hematite-based fuel cell reached an impressive power output of 467 mW cm^{-2} at 600°C [13]. Our latest study further modified the natural hematite through acid treatment, the resulting mineral-based composite exhibited an good ionic conductivity of 0.116 S cm^{-1} at 600°C with an activation energy of 0.50 eV ($400 - 600^\circ\text{C}$) [16]. These preliminary achievements have shown that the use of natural minerals can be an underlying route to developing low-temperature SOFCs (LTSOFCs). Considering the tremendous commercial and practical value of minerals, it is quite necessary to develop more potential from natural materials for SOFC applications.

Recently, a nanocomposite approach has been proposed to prepare a variety of electrolyte materials with desirable ionic conductivities [17–20]. These nanocomposite electrolytes with multiphased ionic conductors exhibited significantly enhanced conductivities compared to conventional electrolytes. A notable example was the composite fabricated from doped ceria and carbonate, such as core-shell structured $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{2-\delta}/\text{Na}_2\text{CO}_3$ (NSDC), which exhibited an excellent ionic conduction property even at extremely low temperatures (over 0.1 S cm^{-1} at 300°C) [21]. Equally significant, another series of nanocomposites comprising of ionic conductor and semiconductor have also been demonstrated as a novel membrane material for SOFCs, showing impressive performances in the temperature range of $400 - 650^\circ\text{C}$ (for instance, the SDC- $\text{Li}_{0.15}\text{Ni}_{0.45}\text{Zn}_{0.4}\text{O}_x$ and NSDC- $\text{Sr}_2\text{Fe}_{1.5}\text{Mo}_{0.5}\text{O}_{3-\delta}$ systems) [22–25]. This kind of material was applied as the membrane layer in SOFCs with a modified architecture, in which the composite was sandwiched by two semiconducting $\text{Ni}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{LiO}_{2-\delta}$ (NCAL) layers [26,27]. On this basis, such a nanocomposite approach provides an underlying strategy for promoting the applications of natural minerals in LTSOFCs.

In the present study, we designed an ionic-semiconducting composite, i.e., hematite/LaCePrO_x-La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3- δ} (hematite/LCP-LSCF), which acted as the intermediate membrane layer for the fuel cell device. The as-designed composite was prepared via a simple and feasible process with materials sourced from robust natural minerals (hematite and LaCePr-carbonate) and perovskite oxide. The phase structure, morphology, and electrical property of the resultant composite are investigated. Then, the composite is evaluated in fuel cell devices at low temperatures ($450 - 600^\circ\text{C}$).

2. Experimental

2.1. Material and preparation

The natural hematite mineral utilized in this study is a high-grade ore exploited from the Laiwu (LW) zone of Shandong province in China [28]. It mainly consists of hematite ($\alpha\text{-Fe}_2\text{O}_3$ 68.359 wt %) and quartz (SiO_2 19.039 wt %) [13]. The LCP-carbonate used in the study was purchased from a rare-earth high-tech company (Baotou, Inner Mongolia, China). It is an industrial-grade mineral composed of lanthanum, cerium, and praseodymium carbonates [15]. The perovskite oxide LSCF was purchased from Sigma-Aldrich, Sweden. The electrode material, commercial NCAL, was obtained from Tianjin Bamo Sci. & Tech. Joint Stock Ltd., China. The nitric acid (HNO_3 , 65%) was obtained from PROLABO-VWR-BDH, France.

The hematite/LCP composite was synthesized through a typical acid-treatment and calcination process, which was reported

previously [16]. The red powder of the hematite/LCP composite was blended with LSCF in an agate mortar at various weight ratios (10 wt %, 20 wt %, 30 wt % and 40 wt % LSCF); it was then wet-ball milled for 12 h using ethanol as the dispersing medium. The resulting mixtures were dried at 300°C and sintered at 700°C for 4 h. Subsequently, the sintered mixtures were adequately ground to form the final product, which is denoted as the hematite/LCP-LSCF composite. For comparative studies, raw hematite (LW) was calcined at 800°C for 4 h, and LCP-carbonate was processed under the same condition to obtain its oxidized product LCP-oxide. In parallel, NCAL-Ni electrode was fabricated using a NCAL slurry and nickel foam (Ni-foam). NCAL was processed into a slurry form by mixing its powder with glycerol in ethanol (10% volume ratio of ethanol) and ground completely. Thereafter, the slurry was pasted evenly on the Ni-foam and desiccated (300°C , 0.5 h) to obtain the NCAL-pasted Ni foam (NCAL-Ni).

2.2. Characterization

The crystal structures of the samples were determined by a Bruker D8 advanced X-ray diffractometer (XRD, Germany, Bruker Corporation) with Cu K α radiation ($\lambda = 1.54060 \text{ \AA}$) as the source operating at 45 kV. The morphologies and microstructures of the samples were investigated by a JSM7100F field emission scanning electron microscope (FESEM, Japan) equipped with energy dispersive spectrometry (EDS) operating at 15 kV. Transmission electron microscopy (TEM) images of the hematite/LCP composite was acquired on a JEOL JEM-2100F field-emission microscope operating at 200 kV.

2.3. Fuel cell construction

The fuel cell devices in this work were assembled by a dry-pressing process. The hematite/LCP-LSCF composite as the membrane layer was sandwiched between two NCAL-Ni electrodes to form a symmetric architecture of NCAL-Ni | hematite/LCP-LSCF | NCAL-Ni. The membrane layer and electrodes were compacted under a pressure of 280 MPa to form a pellet, with an active area of 0.64 cm^2 and a thickness of 1.5 mm. This novel sandwich-shaped device was previously reported in substantial studies [24,26,27]. Note that the NCAL is a new triple ion ($\text{H}^+/\text{O}^{2-}/\text{e}^-$) conducting material, its crystal structure, morphology, and function as an electrode have been intensively investigated [29,30]. The structure diagram and reaction mechanism of our designed fuel cell device are schematically illustrated in Fig. 1.

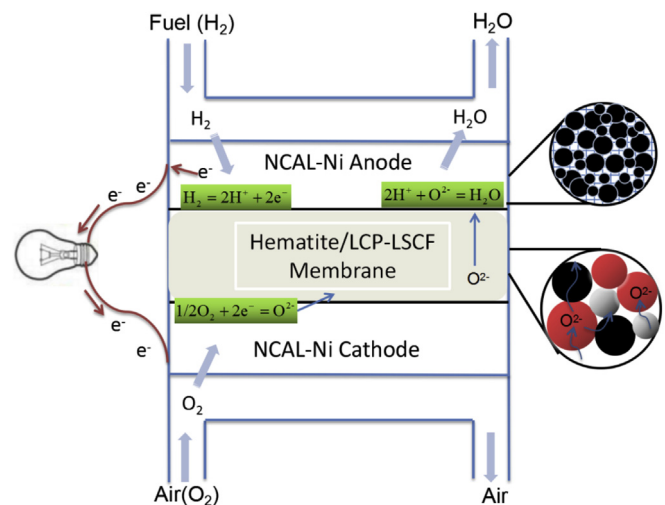


Fig. 1. Schematic diagram of the architecture and mechanism of the fuel cell device.

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