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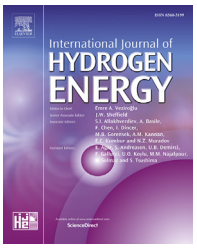

The heterogeneous electrolyte of $CuFeO_2$ nano-flakes composited with flower-shaped ZnO for advanced solid oxide fuel cells

Jing Zhang [1], *Huaibing Song* [1], *Rong Xu*, *Chunjie Yan* [*], *Yan Wu* [**]

Engineering Research Center of Nano-Geo Materials of Ministry of Education, Faculty of Materials Science and Chemistry, China University of Geosciences, 388 Lumo Road, Wuhan, 430074, China

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ABSTRACT

The flower-shaped ZnO was synthesized to form composite with the delafossite structure $CuFeO_2$. The composite heterojunction formed for the ZnO-$CuFeO_2$ composite material demonstrates a profound significance for exploring novel materials in solid oxide fuel cell (SOFC) field. At 550 °C, power outputs of 300 mW cm^{-2} and 468 mW cm^{-2} were achieved for SOFC devices using pure ZnO and composite with $CuFeO_2$ as the electrolytes, respectively. The composite showed a good performance at low temperatures, for instance, it showed a power output of 148 mW cm^{-2} at 430 °C. The studies on photocurrent-time curves with visible light on/off irradiation provided an evidence for electron-hole separation. The heterojunctions separate holes and electrons, preventing short-circuiting while used in the SOFC device. These results demonstrate that introducing the heterojunctions in the electrolyte is an innovative approach for advanced SOFCs.

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Introduction

Today, worldwide industrial development is dependent heavily on fossil fuels. It was also known that the exhaustion and burning of the fossil fuels have brought a series of problems, such as energy consumption and environmental pollution. Therefore, the development of safe, clean and efficient energy is urgently needed [1]. Solid oxide fuel cell (SOFC) as a novel energy conversion device that can convert chemical energy directly into electrical energy has attracted considerable attention [2–4]. The most significant challenge to the current commercialization of SOFC is to cut down its high costs due to the high operating temperature [5]. Decreasing the operational temperature below 600 °C can meet the commercial demands [6–9]. One of the strategies, decreasing the operational temperature, is to seek the new materials possessing of high ionic conductivities at low operating temperatures [10–19].

Currently we found that natural ore has an excellent performance for SOFC at low operational temperatures [20–24]. As an electrolyte, natural hematite and delafossite ($CuFeO_2$) can achieve power outputs of 225 and 421 mW cm^{-2} below 600 °C, respectively [21,22]. Using hematite ore composite with $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) as the electrolyte, the power density can also reach up to 467 mW cm^{-2} at 550 °C [22].

* *Corresponding author.*
** *Corresponding author.*
E-mail addresses: chjyan@cug.edu.cn (C. Yan), wuyan@cug.edu.cn (Y. Wu).
[1] The authors made equal contributions.
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Natural delafossite and yttrium stabilized zirconia (YSZ) composites even achieved a power density of 560 mW cm^{-2} at 550 °C [23]. Using composites of natural delafossite and Li_xZnO-$Sm_{0.2}Ce_{0.8}O_{2-\delta}$ (LZSDC) as the electrolyte, a power density of 637 mW cm^{-2} was realized [21]. The above results indicated that Cu (I)-based delafossite compounds ($CuFeO_2$) have more advantages and better properties for fuel cell to decrease the operating temperatures, owing to its high stability and hole conductivity [25,26]. ZnO is an important wide-bandgap semiconductor applied widely in light emitters [27], piezoelectric transducers [28], phosphors [29], and transparent conducting films [30], which attribute to its high free exciton, native substrate, and wet chemical processing [31]. Owing to its abundance, low-cost, large surface areas, direct electron pathways, and effective light-scattering centers, ZnO is widely used in dye-sensitized solar cell (DSC) applications [32] and SOFCs [33]. Recently ZnO semiconductor was composited with hematite to form a composite electrolyte in advanced SOFCs, which demonstrated promising performances [24].

$CuFeO_2$ is a p-type semiconductor [26,34–36]. By doping, ZnO could obtain n-type conductivity [36–45]. In this work, we composited ZnO with $CuFeO_2$ to construct heterojunctions. The heterojunctions were reported as an effective approach to make charge separation and prevent the electron passing through internally the device thus avoiding the short circuiting problem [10,46]. Besides, nano-redox effective fuel cell reactions can take place on the p and n interfaces, which is helpful to construct the fuel cell. Therefore, composite heterojunctions inside the cell device can effectively make charge separation and prevent the carriers from recombination and generate electricity [10,47]. It gives us the confidence and scientific cognition to construct novel SOFC devices by introducing the heterojunctions as the electrolyte.

Experiment

Preparation of $CuFeO_2$

The $CuFeO_2$ precursor was synthesized by Hydrothermal process [48] and then dried at 70 °C for 2 h. The resulting dried powder was ground in agate mortar and sintered at 300, 600, 900 °C for 2 h at nitrogen atmosphere, respectively.

Preparation of ZnO and ZnO-$CuFeO_2$ composites

The ZnO and ZnO-$CuFeO_2$ composites were synthesized by sol-gel method. All the chemicals used are analytical grade. 0.892 g $Zn(NO_3)_2 \cdot 6H_2O$, 2.118 g $C_6H_5Na_3O_7 \cdot 2H_2O$ and corresponding proportional $CuFeO_2$ were dissolved in 50 mL deionized water at room temperature under magnetic stirring. Then 0.6 g NaOH was added to the solution and stirred for 2 h. After cooling, the gray precipitate formed was filtered, washed with deionized water and absolute alcohol respectively, and dried at 70 °C for 24 h. The as-obtained power was ZnO-$CuFeO_2$ composite. For different weight ratios of ZnO-$CuFeO_2$ composites, ZnO:$CuFeO_2$ = 95:5, ZnO:$CuFeO_2$ = 90:10, ZnO:$CuFeO_2$ = 85:15, ZnO:$CuFeO_2$ = 80:20, we named them as ZC9505, ZC9010, ZC8515, ZC8020. The pure ZnO was prepared similarly without the addition of $CuFeO_2$ by following the above procedures.

Characterization of materials

The phase structure of samples was analyzed by D8-FOCUS X-ray diffractometer with Cu Kα radiation source at a wavelength of 0.15418 nm (Bruker, Germany). The microstructure of the samples was analyzed using a field emission scanning electron microscope (FESEM, JEOL JSM 7100F Japan). Transmission electron microscopy (TEM) and high resolution transmission electron microscopy (HRTEM) were performed with a FEI TECNAI G2 F30 to get a clear vision of heterojunctions of $CuFeO_2$ and ZnO. Commercial $Ni_{0.8}Co_{0.15}Al_{0.05}Li$ oxide (NCAL, Tianjin Baomo Joint Hi-tech Venture, China), was directly used for preparation of pellets. Through pulping, NCAL was coated on one side of Ni-foam to prepared electrode.

Fabrication and measurement of fuel cells

In this study, a novel SOFC device was fabricated as a sandwich in which the ZnO-$CuFeO_2$ composite was placed with NCAL-coated Ni-foam electrodes on both sides, i.e., Ni-NCAL/ZnO-$CuFeO_2$ composite/NCAL-Ni. All the samples, with a weight of 0.3 g, were compressed into a pellet under pressure of 200 MPa, respectively. The pellets have a uniform active area (0.64 cm^2) and thickness (2 mm). The devices were sintered from its ambient value to 550 °C and held for about 30 min before carrying out the electrochemical measurements. The performance of the cell, the *I-V* (current density-voltage) and *I-P* (current density-power density) curves, were recorded on a fuel cell tester (ITECH DC ELECTRONIC LOAD, IT8511) with the temperature range from 430 to 550 °C. The electrochemical impedance spectra (EIS) of the pellet was measured by CHI660B electrochemical workstation (Chenhua, China) in the same temperature range. Pure hydrogen gas was fed as fuel into the "anode" side at a rate of 110 mL/min while air was used as the oxidant with a flow rate of 100 mL/min. Each side of the cell was maintained at the pressure of 1 atm. The DC polarization test was prepared (using platinum sheet as the electrode and with an active area of 0.64 cm^2 and 1 mm thickness) by ITECH DC Power Supply (IT6861A) at N_2 atmosphere.

Photocurrent and Hall effect tests

The photocurrent tests were performed by CHI660B electrochemical workstation (Chenhua, China). The tests were measured via a three-electrode arrangement, the counter electrode (Pt wire), the reference electrode (an Ag/AgCl electrode with a concentration of 3.0 M KCl), and the working electrode (a small piece of glass coated with an In-doped SnO_2 conductive substrate). The sample was mixed with alcohol and polyethylene glycol into slurry and deposited on the side

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