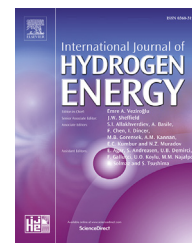




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## Remarkable ionic conductivity and catalytic activity in ceramic nanocomposite fuel cells

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### ABSTRACT

Although ceramic nanocomposite fuel cells (CNFCs) have attracted the attention of the fuel cell community due to their low operating temperature (<600 °C), often the performance of the cells is limited due to the low ionic conductivity of the electrolyte and the sluggish reaction kinetics at the electrodes. This results in high ohmic and charge transfer losses in the cell performance. Here we report nanocomposite electrolyte (GDC-NLC) and electrodes (NiO-GDC-NLC and LSCF-GDC-NLC as anode and cathode respectively) with enhanced ionic conductivity and catalytic activity respectively, which significantly improve the ionic transport in the electrolyte layer (ohmic losses  $\approx 0.23 \Omega \text{ cm}^2$ ) and the reaction kinetics at the electrodes (polarization losses  $\approx 0.63 \Omega \text{ cm}^2$ ). Microstructural and phase changes in the materials were characterized with X-ray diffraction, scanning electron microscopy, and differential scanning calorimetry to understand the mechanisms in the cells. Our button fuel cell produced an outstanding performance of  $1.02 \text{ W/cm}^2$  at 550 °C.

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### Introduction

Conventional ceramic fuel cells, also known as solid oxide fuel cell (SOFC) and molten carbonate fuel cell (MCFC) are fuel cell technologies that offer many new possibilities. As a result of the limited ionic conductivity of the most commonly used yttria stabilized zirconia (YSZ) based electrolytes in SOFC, these cells operate at high temperatures of 800–1000 °C [1,2]. This high-temperature operation can cause several degradation issues in the device, material problems, and challenges for the commercialization of this technology [3,4]. Alternative electrolytes based on doped ceria have been successfully used to reduce the operating temperature of the cell to 550 °C. These cells are called low-temperature solid oxide fuel cells

(LT-SOFCs) [5]. However, LT-SOFC suffers from ohmic and activation losses primarily due to limited oxygen ion transport through the electrolyte and slow reaction kinetics at the electrodes, respectively [6].

MCFC is also a high-temperature fuel cell (~650 °C) that uses an electrolyte consisting of alkali carbonates mixtures that are mostly eutectic, and a chemically inert porous ceramic matrix of beta-alumina [7,8]. At operating temperature, the corrosive molten alkaline carbonates can degrade the NiO cathode in a reducing atmosphere [9–11]. Generally, for commercialization of a fuel cell technology, stability for 40,000 h is required. It is thus essential to improve the stability of the MCFC either by decreasing the operating temperature or by replacing the cell components with more stable ones.

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Ceramic nanocomposite fuel cells (CNFCs) are developing as a potential hybrid fuel cell technology that uses potential materials from both the SOFC and MCFC to provide the more efficient device at a lower temperature (400 °C–600 °C) [12,13]. The foundation of these composite electrolytes were laid down already almost 2 decades ago [14–16]. In a comparative study of different ceria-salt composite materials in IT-SOFC electrolytes, the doped ceria – alkali carbonates performed with the highest power density (800 mW/cm<sup>2</sup>) [15]. Further progress in the development of the composite materials have been resulting in improved fuel cell devices, which is thoroughly covered in the literature [17–26]. The nanocomposite cells offer high ionic conductivity of the electrolyte at low temperatures (550 °C) [23,27–29] which overcome ohmic losses and provide more efficient and stable electrode options [12,25,30]. Mostly, these fuel cells utilize nanocomposite electrolyte mostly consisting of doped ceria and a eutectic mixture of binary or ternary alkali carbonates [12,31,32]. These fuel cells manifest multipolar ions transport through the electrolyte taking advantage of the different channels available for their conduction [13,33,34]. There are four different channels available for ions conduction: through the oxide phase, through the molten phase, through the interfaces between the oxide phase and molten phase, and their combination. It is suggested [13,33] that the conduction through the molten phase and through a combination of molten phase and interfaces dominate the conduction process in the electrolyte at operating temperature above the eutectic temperature, ~500 °C in case of Na<sub>2</sub>CO<sub>3</sub> and Li<sub>2</sub>CO<sub>3</sub> eutectic mixture. At low temperatures, less than 400 °C in case of binary electrolyte, the conduction is dominated through the oxide phase. Conduction through the interfaces dominates between 400 °C and 500 °C temperature region. Fig. 1 shows the native (Li<sup>+</sup>, Na<sup>+</sup>, CO<sub>3</sub><sup>2-</sup>) and foreign (O<sup>2-</sup>, OH<sup>-</sup>, H<sup>+</sup>) ion species in the binary nanocomposite electrolyte [35].

The application of the nanocomposite materials in novel and intriguing concept of a single component fuel cell or semi-ionic device have resulted in breakthrough in the fuel cell research [36–39]. In traditional 3-layer cell structures, a wide range of performances of the CNFCs have been reported in the literature which varies from 60 mW/cm<sup>2</sup> to around 1 W/cm<sup>2</sup>. The variation in the performances can be attributed to the handling of the materials during their synthesis and the fuel cell fabrication process. Furthermore, accuracy of the measurement setup is critical to the authenticity of the measurements. With careful handling of the cells during all the

fabrication and measurement steps, our CNFCs achieved a high power density of 1.02 W/cm<sup>2</sup> at 550 °C, which is significantly higher than the power density reported in several studies in the literature: 92 mW/cm<sup>2</sup> at 550 °C [40], 60 mW/cm<sup>2</sup> at 600 °C [41], 490 mW/cm<sup>2</sup> at 550 °C [42], and is comparable to other high performances reported in the literature: 800 W/cm<sup>2</sup> at 650 °C [15], 800 mW/cm<sup>2</sup> at 550 °C [43], 590 mW/cm<sup>2</sup> at 600 °C [44].

## Experimental section

### Materials and fabrication

The materials and chemicals that were used for the preparation of nanocomposite electrolyte and electrodes were purchased from Sigma-Aldrich. Here is a list of the chemicals: gadolinium doped ceria (GDC), sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>), nickel oxide (NiO), lanthanum strontium cobalt ferrite (LSCF), polyvinyl alcohol (PVA) and ethyl cellulose (EC).

The electrolyte powders were prepared by making a eutectic mixture (57:43 wt%) of Na<sub>2</sub>CO<sub>3</sub> and Li<sub>2</sub>CO<sub>3</sub> (NLC) through solid route method and then mixing binary carbonate (25 wt%) and GDC (75 wt%) in a planetary ball milling machine at 120 RPM for 1 h using acetone as the media. Then the powders were dried and grinded with a mortar and a pestle. The nanocomposite electrolyte pellets were prepared by placing the electrolyte powder into a 13 mm die and pressing at 250 MPa for 2 min. The pellets were sintered at 690 °C for 1 h. Gold paste was painted on respected sides of the pellets to improve the electrical contact between pellets and gold current collector wires.

The electrode powders were prepared through solid route method and ball milling similarly to electrolyte powders. The anode powder consisted of NiO, electrolyte material, PVA and EC in weight % ratio 45:45:5:5 respectively. The cathode powder consisted of LSCF, electrolyte material, PVA and EC in weight % ratio 45:45:5:5 respectively. The CNFCs were prepared by sequentially placing Ni foam, anode powder, electrolyte powder, cathode powder and another Ni foam into a 13 mm die and pressing at a pressure of 250 MPa for about 2 min. The pellets were sintered at 690 °C for 1 h and gold paste was painted on the respected sides of the pellets to achieve improved electrical contacts in the measurement setup.

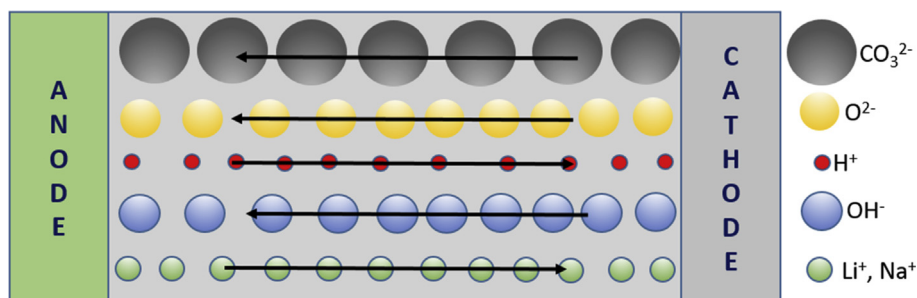


Fig. 1 – Schematic presentation of ionic transport in the ceramic nanocomposite electrolyte.

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