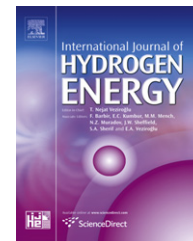


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Technical Communication

Validation of H^+/O^{2-} conduction in doped ceria–carbonate composite material using an electrochemical pumping method

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ABSTRACT

A hydrogen and oxygen electrochemical pump technique has been employed to elucidate the conduction of proton and oxygen ion in a doped ceria–carbonate composite electrolyte for intermediate temperature solid oxide fuel cells. The composite material shows efficient conductivities of both of the two ions at 650 °C. The molten carbonate phase is important for the migration of both of the two ions. The mechanism of the conduction of proton and oxygen ion is also discussed.

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1. Introduction

Intermediate temperature (500–800 °C) solid oxide fuel cells (ITSOFCs) have attracted much attention in recent years for their advantages in operating cost and material selection compared to the conventional high temperature SOFCs [1]. A key requirement of the ITSOFCs is to develop an electrolyte material with high ionic conductivity at the IT range [2]. A sort of doped ceria (DCO)-carbonate composite materials shows a promising behavior and has been intensively investigated in the past two decades [3,4]. The composite electrolyte presents

a high overall ionic conductivity ($>0.1 \text{ S cm}^{-1}$) above the melting point of the carbonate (400–500 °C) while a single cell based on this material gives a high performance in IT range, e.g. 1700 mW cm^{-1} at 650 °C [5,6].

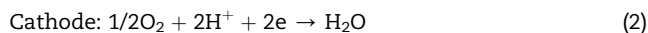
It has been proposed that a H^+/O^{2-} binary conduction existing in the composite electrolyte leads to the high ionic conductivity and excellent cell performance [7]. This mechanism was supported mainly on the experimental fact that the water was condensed in the outlet gases of both the anode and cathode sides during the cell operation. Reactions (1)–(4) can be written to account for the ion electrode reactions.

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In the case of H^+ conduction:



In the case of O^{2-} conduction:



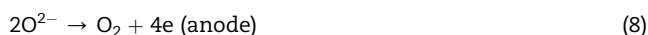
However, the formation of water in both the electrode chambers could be also caused due to the leakage of the reactants through the composite electrolyte, i.e. the non-electrochemical reaction of hydrogen and oxygen of leaked through. Recently, efforts have been made to prove the existence of O^{2-} and H^+ transfer using the AC impedance and four probe techniques under H_2 and oxygen atmospheres [8,9]. It has been reported that the conductivity of the composite electrolyte under H_2 atmosphere presents a higher value than in the oxidant atmosphere, which indicates the existence of proton conduction. However, such a change of conductivity is also possibly attributed to the reduction of Ce^{4+} to Ce^{3+} which increases the electronic conductivity [10]. Thus, a more reliable method is needed to carefully measure the O^{2-} and H^+ conduction in the composite material.

The electrochemical pumping technique has been widely used for gas separation, e.g. H_2 or O_2 , using a proton conductor or an oxide ionic conductor, respectively [11,12]. The schematics of the hydrogen and oxygen electrochemical pumps are shown in Fig. 1. With the application of a current/potential, H_2 or O_2 is selectively pumped from one side of the ionic conductor to the other side with the involvement of the electrochemical reactions (5)–(8) on the surface:

For the hydrogen electrochemical pump



For the oxygen electrochemical pump



In this work, hydrogen and oxygen electrochemical pumps are first used to confirm and investigate the conduction of H^+ and O^{2-} in the samarium doped ceria (SDC)– $(Li_{0.52}Na_{0.48})_2CO_3$ composite material. A galvanic step is applied to the composite material under ambient pressure using the electrochemical pumping configuration. The O_2 or H_2 is supplied to one side of the reactor while the products are measured by a mass spectrometer on the other side during the polarization. The results prove the existence of the conduction of H^+ and O^{2-} directly. The pathways of both of the ions are also proposed.

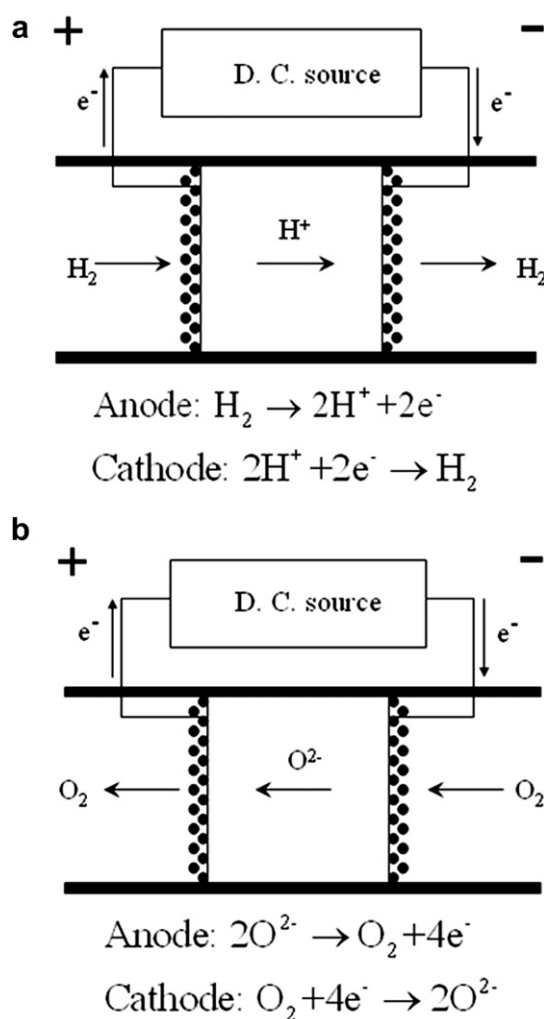


Fig. 1 – The principle of the hydrogen (a) and oxygen (b) electrochemical pumps.

2. Experimental

The preparation of the SDC– $(Li_{0.52}Na_{0.48})_2CO_3$ composite electrolyte has been described in our previous work [6]. The weight ratios of the composites (SDC: carbonate) were chosen to be 95:5, 90:10, 80:20 and 70:30, which were marked as SDC-5LN, SDC-10LN, SDC-20LN and SDC-30LN, respectively. The composite electrolyte powder was pressed at 300 MPa into a cylindrical pellet with a diameter of 13 mm and a thickness of 1 mm using a uniaxial die-pressing technique. The green pellets were then sintered at 650 °C for 1 h. Platinum electrodes were subsequently prepared by painting platinum paste onto each side of the pellet.

The procedure for the hydrogen pump measurement was the same as that illustrated in Fig. 1(a). Dry hydrogen was supplied to the anode, and dry argon was used as the carrier gas in the cathode side. The gas flow rates in both sides were 100 ml min⁻¹ (STP). The hydrogen electrochemical pump was operated at 500–650 °C. A direct current was sent to the cell by an electrochemical workstation (VersaSTAT 3, Ametek). To determine the Ohm drop of the voltage on the electrolyte membrane, a current interruption method was employed,

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