

Communication

Hybrid-solid oxide electrolysis cell: A new strategy for efficient hydrogen production

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ABSTRACT

Water electrolysis based on a solid oxide electrolysis cell (SOEC) has potential to be cost-effective, environmentally friendly, and highly efficient for hydrogen production. There are two types of SOECs, depending on electrolyte materials: oxygen ion conducting SOECs (oxygen-SOECs) and proton conducting SOECs (proton-SOECs). Here we report our new findings in exploring a SOEC based on a mixed-ion conductor that can transport both oxygen ion and proton at the same time, which is denoted as “Hybrid-SOEC”. When $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.1}\text{Yb}_{0.1}\text{O}_{3-\delta}$ was used as an electrolyte, the Hybrid SOEC shows the highest efficiency, demonstrating a current density of 3.16 A cm^{-2} at 1.3 V and 750°C in 10% humidified hydrogen at hydrogen electrode and 10% humidified air at air electrode. Moreover, the Hybrid SOEC exhibits no observable degradation in performance for more than 60 h of continuous operation, implying a robust system for hydrogen production.

1. Introduction

Environment-friendly and clean fuels have attracted world-wide attention due to the growing concerns about global warming and other environmental issues associated with the heavy consumption of fossil fuels [1]. Hydrogen is considered a promising alternative to fossil fuels because it is the cleanest fuel on the earth and has the highest gravimetric energy density among all chemical fuels (143 kJ kg^{-1}) [2,3]. However, hydrogen does not exist naturally on the earth and must be produced from water or hydrocarbons. In fact, most hydrogen used today (> 90%) is produced from hydrocarbons by the steam reforming, resulting in greenhouse gas (CO_2) emission and high cost [4]. Other environmentally benign hydrogen production processes include photoelectrochemical, thermochemical, and electrochemical water splitting. Among them, water electrolysis is the most viable approach when renewable energy sources can be used to run the process. Low temperature electrolysis such as polymer electrolyte membrane (PEM) electrolysis and alkaline water electrolysis ($< 100^\circ\text{C}$), however, needs significant amount of the electricity and noble metal-based electrocatalysts, leading to high cost and low energy efficiency [5]. In contrast,

high-temperature steam electrolysis based on solid oxide electrolytic cells (SOECs) is much more efficient because of their advantages for the water splitting both thermodynamically and kinetically [6].

Oxygen-SOEC is one of the most promising candidate for the high temperature water electrolysis [7]. In this system, the hydrogen is produced from the water-electrolysis reaction at the hydrogen electrode side as described in Fig. 1(a). As an electrolyte material, yttria-stabilized zirconia (YSZ) is conventionally used because the YSZ exhibits sufficient pure oxygen ion conductivity at high temperatures ($> 800^\circ\text{C}$) [8–11]. Despite the benefits of high temperature operation, however, wide application of the oxygen-SOEC has been inhibited because of critical issues (e.g., poor long-term cell stability, interlayer diffusion, fabrication and materials problems).

For these reasons, proton-SOECs using proton conducting oxides (PCOs) as electrolyte materials have acquired much interests because of their intermediate to low temperature operation [12]. In the proton-SOEC system (Fig. 1(b)), the proton is generated by water electrolysis at the air electrode and this proton permeates through the pure proton-conducting electrolyte, and the permeated proton reacts with electrons to produce hydrogen at the hydrogen electrode. Recently, extensive

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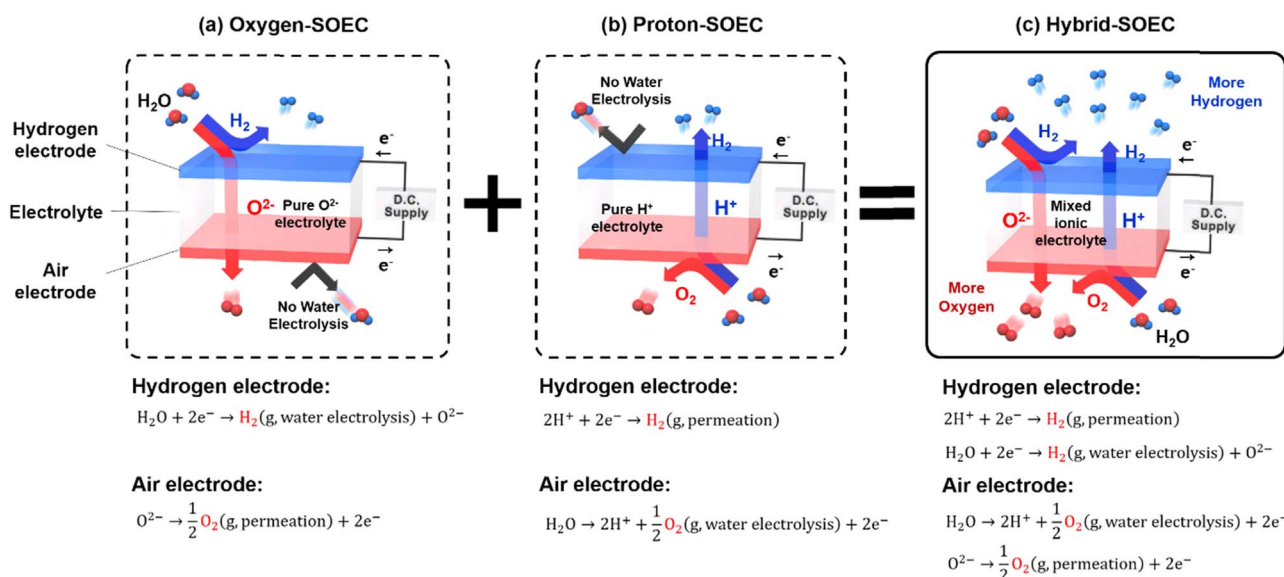
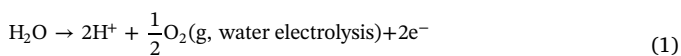


Fig. 1. Schematic diagrams of the working principle. (a) Oxygen-SOEC, (b) proton-SOEC, and (c) Hybrid-SOEC system. In the oxygen- and proton-SOEC systems (a and b), even in the presence of steam on both electrodes, the electrochemical reaction for the water-electrolysis can occur at only one electrode. On the contrary, when mixed ion conducting electrolyte is used, the water electrolysis can occur at both at both air and hydrogen electrodes (so-called Hybrid-SOEC operating system), because the electrolyzed ion species (O^{2-} and H^+) can be counter-diffused across the mixed ionic conducting electrolyte. Consequently, in the Hybrid-SOEC system (c), more hydrogen could be generated by allowing simultaneous the oxygen-SOEC and proton-SOEC operation.

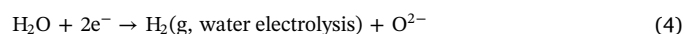
efforts have been devoted to developing the PCOs based on barium-zirconates (BZO) and barium-cerates (BCO) to improve their chemical stability and proton conductivity. Among those PCOs, aliovalent-doped $\text{BaCe}_{1-x}\text{Zr}_x\text{O}_{3-\delta}$ ($0 < x < 1$) have demonstrated high proton conductivity and successfully eliminated major issues such as poor sinterability and low proton conductivity [13,14]. Even with these advantages of BZO and BCO, the efficient performance based on zirconates/cerates electrolyte has been limited mainly due to the large polarization resistance of the air electrode. Recently, we reported that layered perovskite material, $\text{NdBa}_{0.5}\text{Sr}_{0.5}\text{Co}_{1.5}\text{Fe}_{0.5}\text{O}_{5+\delta}$ (NBSCF), exhibits remarkable electro-chemical performance as an air electrode based on the barium-cerate electrolyte compared to other conventional electrodes because of its triple-conducting property ($\text{O}^{2-}/\text{H}^+/\text{e}^-$) [15].

One of the notable points from these operating principles is that the water-electrolysis reaction can theoretically be occurred in both air electrode and hydrogen electrode depending on the type of ions (O^{2-} or H^+) that permeates the electrolyte. However, the water-electrolysis reaction is limited to only one side of electrode in the conventional SOECs, because only one ion can permeate across the single ion conducting electrolyte (e.g., YSZ, BCOs, and BZO) even though steam is fed to both electrodes of the SOEC. In this regard, the introduction of the electrolyte conducting simultaneously both O^{2-} and H^+ species could be a good approach for the efficient hydrogen and oxygen production. In this system, water electrolysis can be occurred at both air and hydrogen electrodes (so-called Hybrid-SOEC operating system, Fig. 1(c)) because the electrolyzed ion species (O^{2-} and H^+) can be counter-diffused in the opposite direction across the mixed ionic conducting electrolyte. In other words, in the Hybrid-SOEC system, more hydrogen and oxygen could be generated simultaneously from the permeated proton (Eqs. (1) and (3)) and water-electrolysis (Eqs. (2) and (4)). The overall reactions at the air and hydrogen electrode in the Hybrid-SOEC system can be expressed as follows:

Air electrode:



Hydrogen electrode:



In this study, for the first time, we demonstrate the novel concept of “Hybrid-solid oxide electrolysis cell” (Hybrid-SOEC) based on the mixed ionic conducting electrolyte. To realize the efficient and stable Hybrid-SOEC operation, we adopt the $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.1}\text{Yb}_{0.1}\text{O}_{3-\delta}$ (BZCYyb) as an electrolyte because this material has both protonic and oxide ion defect, resulting in high mixed ionic (O^{2-} and H^+) conductivity under proper condition [16]. For electrode materials, we choose $\text{NdBa}_{0.5}\text{Sr}_{0.5}\text{Co}_{1.5}\text{Fe}_{0.5}\text{O}_{5+\delta}$ (NBSCF) - BZCYyb composite and Ni-BZCYyb composite as an air electrode and hydrogen electrode, respectively. The NBSCF layered perovskite can conduct protons, oxygen ions, and electrons, leading to increased number of electrochemically active sites on the entire surface of the air [15,17]. These properties of the NBSCF may improve the catalytic activity for steam dissociation and O_2 evolution. Also, Ni shows excellent electrocatalytic activity for water splitting and hydrogen evolution and good compatibility with making Ni-BZCYyb composite as an ideal hydrogen electrode for the Hybrid SOEC [18]. The Hybrid-SOEC exhibits outstanding electro-chemical performance with stability compared to other SOECs and representative water-electrolysis devices reported in the literatures.

2. Results and discussion

In many cases, reactions between the electrode and the electrolyte materials may result in insulating phases at the interfaces, which blocks ionic and electronic transport across the interface between the electrode and the electrolyte [19]. Fig. S1 shows XRD patterns of the NBSCF-BZCYyb composite fired at 950°C for 4 h. All the peaks correspond to the characteristic peaks for either the NBSCF or the BZCYyb, indicating that there are no obvious interfacial reactions or distinct secondary phases between them.

The schematic diagram and experimental details for Hybrid-SOEC measurements are described in Fig. S2. Hydrogen was supplied to the hydrogen electrode side while air was supplied to the air electrode side with a flow rate of 100 mL min^{-1} . In Hybrid-SOEC measurements, 10% steam was supplied to both electrodes from a water bubbler kept at a constant temperature for the required amount of steam and the tubes

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