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Improvements in co-electrolysis performance and long-term stability of solid oxide electrolysis cells based on ceramic composite cathodes

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ABSTRACT

The co-electrolysis of CO₂ and H₂O was performed using a solid oxide electrolysis cell with yttria-stabilized zirconia (YSZ) as the electrolyte and a La_{0.7}Sr_{0.2}Cr_{0.5}Mn_{0.5}O₃ (LSCM)-YSZ composite as the cathodic material. The LSCM-YSZ composite cathode showed better co-electrolysis performance than did a La_{0.7}Sr_{0.3}V_{0.9}O₃ (LSV)-YSZ composite cathode, suggesting that the catalytic activity of LSCM is higher than that of LSV. After small amounts of CeO₂ and Pd had been added as catalysts to the two composite electrodes, the LSCM-YSZ cathode showed better co-electrolysis performance than that of the LSV-YSZ composite electrode. The long-term stability of the co-electrolysis performance of the LSCM-YSZ composite cathode was higher than that of the LSV-YSZ composite cathode. These results show that the electrolysis performance of composite cathodes containing highly active materials such as CeO₂ and Pd as catalysts can be improved further by choosing the proper base material.

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Introduction

In order to reduce the greenhouse effect, it is necessary to be able to capture and store CO₂ safely. Of late, a novel technology for recycling stored CO₂ in the form of carbon sources, called carbon capture and reuse, is being studied actively [1,2]. Recent efforts toward reusing stored CO₂ have focused on the electrochemical conversion of CO₂ into a hydrocarbon fuel

through the use of solid oxide electrolysis cells (SOECs) [2–4]. In addition to allowing the recycling of CO₂, SOECs can also be used to store energy from renewable sources such as solar energy, wind power, and geothermal power in the form of chemical energy such as hydrogen or CO [5,6].

In particular, the co-electrolysis of CO₂ and H₂O at high temperatures has received much interest because it can directly generate syngas, a mixture of CO and H₂ that can be converted into a liquid hydrocarbon fuel through the

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Fischer–Tropsch catalytic process [6–8]. During the co-electrolysis process, the oxygen ions that originate from CO₂ or H₂O at the cathode of the SOEC after the application of a current are transported to the oxygen electrode, resulting in the synthesis of syngas at the cathode [6,9].

Recently, syngas was generated successfully through co-electrolysis performed using a SOEC based on electrodes made from a Ni-yttria-stabilized zirconia (YSZ) cermet [7]. The long-term stabilities of these electrodes under CO/CO₂ atmosphere were also measured [10]. During operation at 1123 K, for a current of 0.5 A cm⁻², the voltage increased at a rate of approximately 0.5 mV h⁻¹; the sulfur poisoning of the Ni present in the electrodes was suggested as the likely reason for the performance degradation [10]. Although various factors affecting composite electrode performance, such as carbon coking, sulfur poisoning, and pressurization, have been investigated extensively, most of these studies have been performed using Ni-YSZ cermet-based cathodes [10–12]. Therefore, the effects of catalysts other than Ni on the co-electrolysis performance of YSZ composite electrodes and their long-term stability remain unknown.

On the other hand, in the case of fuel cells, there have been intensive efforts to replace the Ni in the electrodes with ceramics that are electronically conductive under reducing conditions. A number of different ceramics such as (La_{0.8}Sr_{0.2})(Cr_{0.5}Mn_{0.5})O₃ (LSCM), (La_{0.7}Sr_{0.3})VO₃, and La_{0.3}Sr_{0.7}TiO₃ have been investigated to determine their effects on the ohmic resistance and the degree of electrode polarization as well as their chemical stabilities against carbon coking and sulfur poisoning [13–15]. In particular, LSCM exhibits higher oxygen diffusivity than does La_{0.8}Sr_{0.2}MnO₃ (LSM). It also exhibits good fuel cell performance, even in the absence of a catalyst, at temperatures as high as 1173 K [16,17].

Recently, a (La_{0.7}Sr_{0.3})V_{0.9}O₃ (LSV)-YSZ ceramic composite was employed as the cathodic material during co-electrolysis, and the effects of CeO₂ and Pd as catalysts on the co-electrolysis mechanism were investigated [9]. An LSCM-based electrode containing CeO₂ and Pd as catalysts was used to electrolyze dry CO₂ in the absence of steam. The co-electrolysis performance of LSCM/Gd_{0.1}Ce_{0.9}O_{1.95} (GDC) composite electrode that did not contain any catalysts has also been studied [18,19]. However, the effect of ceramic electrode composition on their co-electrolysis performances has not yet been elucidated, and the long-term stability of such ceramic composite electrodes remains unknown.

In this work, we used LSV- and LSCM-based ceramic composite cathodes for co-electrolysis and studied their performances as well as their long-term stabilities, comparing the two materials. We also tried to determine the critical parameters that determine the co-electrolysis performance and long-term stability of cathodes based on these materials, in order to improve their performances.

Experimental

La_{0.8}Sr_{0.2}FeO₃ (LSF) was used for the air electrode and the tape casting and infiltration method were used to make the target SOECs: LSF-YSZ|YSZ|LSV-YSZ and LSF-YSZ|YSZ|LSCM-YSZ. A detailed description of the fabrication process can be found

elsewhere [14]. The tapes for the dense YSZ electrolyte and the porous YSZ scaffold were fabricated through the tape casting process using a slurry composed of an organic solvent, a dispersant, a binder, and a plasticizer. For the porous YSZ scaffold, graphite powder was added to the slurry to act as a pore former. The tape for the porous scaffold was laminated on both sides of the tape for the dense YSZ electrolyte. This was followed by sintering at 1773 K for 4 h. The thicknesses of the dense electrolyte and the porous scaffold were approximately 80 μm and 40 μm, respectively. And the diameters of the cells and the electrodes were 11 mm and 7 mm.

Aqueous solutions of precursors of La_{0.8}Sr_{0.2}FeO₃ (LSF), LSV, and LSCM were prepared by dissolving the appropriate amounts of the raw materials in water. The raw materials were La(NO₃)₃·6H₂O (99.9%, Alfa Aesar), Sr(NO₃)₂ (99.9%, Alfa Aesar), NH₄VO₃ (99%, Alfa Aesar), Fe(NO₃)₃·9H₂O (99.6%, Alfa Aesar), Cr(NO₃)₃·9H₂O (99.3%, Alfa Aesar), Mn(NO₃)₂·xH₂O (99.9%, Alfa Aesar), and citric acid. The number of moles of citric acid in each aqueous solution was equal to the number of moles of the cations in the solution.

To fabricate the LSF-YSZ, LSV-YSZ, and LSCM-YSZ composite electrodes, the aqueous solutions of precursors of LSF, LSV, and LSCM, respectively, were infiltrated into the porous scaffold several times until the target volume fraction of the electrode materials had accumulated in the scaffold. LSF (20 vol%) was loaded onto the air electrode while either LSV (15 vol%) or LSCM (30 vol%) was loaded onto the cathode. Because the electronic conductivity of LSCM is lower than those of LSF or LSV, the amount of LSCM loaded was larger than those of LSF and LSV.

After the target loading rate had been achieved, the composite electrodes were fired. The final calcination temperatures of the LSF, LSV, and LSCM electrodes were 1123 K, 973 K, and 1223 K, respectively. For comparison, the LSCM electrode was also fired at 1473 K, which is the widely used calcination temperature for LSCM electrodes fabricated using the infiltration method [13]. Finally, the catalysts, which were CeO₂ (2.8 vol%) and Pd (0.5 vol%), were added to the cathodes by making a CeO₂ precursor solution (added as 1 M Ce(NO₃)₃, Alfa Aesar, 99.5%) and a Pd solution (0.45 M tetraamminepalladium (II) nitrate solution, Alfa Aesar, 99.9%), respectively, infiltrate the cathodes. The fabricated cells having the LSCM-YSZ and LSV-YSZ composite cathodes, which contained CeO₂ and Pd as catalysts, are hereafter referred to as the LSCM-cell and the LSV-cell, respectively.

To assess the cell performances, a Ag paste was painted on the top of the electrodes and the resultant Ag layer was used as the current collector. The fabricated cell was mounted on an alumina tube using a ceramic adhesive (Ceramabond 552, Aremco). Humidified (3% H₂O) air was made to flow into the air electrode side (the sweep side) of the cells, while different reducing gases representing the four different operational modes were made to flow into the cathode sides (the feed sides): humidified (3% H₂O) hydrogen to simulate fuel cell operation, humidified (30% H₂O) 4% H₂/balance Ar to simulate steam electrolysis, dry 20% CO/balance CO₂ (CO/CO₂) to simulate CO₂ electrolysis, and humidified (6% H₂O) CO/CO₂ to simulate co-electrolysis. The steam content in the CO/CO₂ mixture for the co-electrolysis operation was chosen to be 6% H₂O, in keeping with a previous work [9].

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