§2. A Study for Hydrogen Isotope Separation and Sensing using Proton Conducting Oxide

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Introduction

Nuclear fusion utilizes hydrogen isotope as a fuel, and thus the efficient methods for hydrogen isotope sensing and separation have to be developed. This study has dealt with hydrogen separation using electrochemical hydrogen pumps and steam electrolysis. Proton conducting oxide is used for this purpose. In addition, electrolyte and electrode are investigated for the development of highly sensitive and selective hydrogen isotope sensors. Through these researches, establishment of hydrogen isotope separation and sensing methods will be directed.

Proton conduction occurs in ABO_3 perovskites to which aliovalent cation doping is conducted; typically A=Ca, Sr, Ba, B=Ce, Zr, and In, rare earth element as dopant. Particularly for the application to steam electrolysis $SrZr_{0.5}Ce_{0.4}Y_{0.1}O_{3-\alpha}$ (α is the molar amount of oxygen vacancy caused by the aliovalent cation doping and incorporation of water molecules into the oxide ion vacancies) has been the optimized composition: denoted below as SZCY541 [1]. In this work, a thin film of SZCY541 was fabricated on the substrate made of NiO/SZCY541composite to reduce the ohmic resistance and hence the energy efficiency. Steam electrolysis performance was examined for the thin-film cell.

Experiment

$SrCO_3$, ZrO_2, CeO_2, Y_2O_3 were used as starting materials to prepare SZCY541. The appropriately weighed amounts were mixed and fired in air at 1200°C. The obtained SZCY541 powder was mixed with NiO powder by a ball mill, pressed into pellet shape and fired at 1200°C to obtain a cathode substrate in pellet form with 14 mm diameter and 0.5 mm thickness. Another portion of the SZCY541 powder was immersed in ethanol dispersant and ethyl cellulose (as a binder) and ball milled to obtain precursor paste. The obtained paste of SZCY541 was screen printed on to the NiO/SZCY541 cathode substrate and co-fired at 1400°C to prepare thin SZCY541 layer on the substrate. The thickness of the electrolyte layer $Sm_{0.5}Sr_{0.5}CoO_3$ was about 20 micron. A paste of $Sm_{0.5}Sr_{0.5}CoO_3$ was hand painted as the anode in 6-mm-diameter circle on the surface of SZCY541 thin film (opposite to the substrate). The specimen was attached to the electrochemical cell housing (NorECs ProboStat) with Pylex glass gasket and heated at 950°C to obtain the final chemical cell of steam electrolysis. The cell was the kept at 600°C and the cathode substrate was reduced by 20% Hydrogen (Ar base plus moisture) to prepare Ni/SZCY541 cermet, in which nickel took the form of porous body. Steam at 20% diluted with Ar gas was supplied to the anode at 600°C and a direct current was sent to the cell. Hydrogen generated at the cathode was swept on Ar gas containing 1% hydrogen and 2% moisture. Hydrogen generation was evaluated by gas chromatography and overvoltage character was measured by a current interrupt method, respectively.

Results and Discussion

SEM observation confirmed a 20-micron film of SZCY541 formed on the NiO/SZCY541 substrate. In the experiment of steam electrolysis, 100% current efficiency was achieved up to the current density 100 mA/cm^2, i.e., the rate of hydrogen generation determined by gas chromatography and the flow rate of the cathode sweep gas agreed with that calculated from the Faraday law. At the current density of 100 mA/cm^2, the overvoltage consisted of 0.18 V by ohmic loss and 0.24 V by the electrode overpotentials, both being satisfactorily low. This result suggests that the present steam electrolysis cell can well extract hydrogen from water molecule electrochemically.

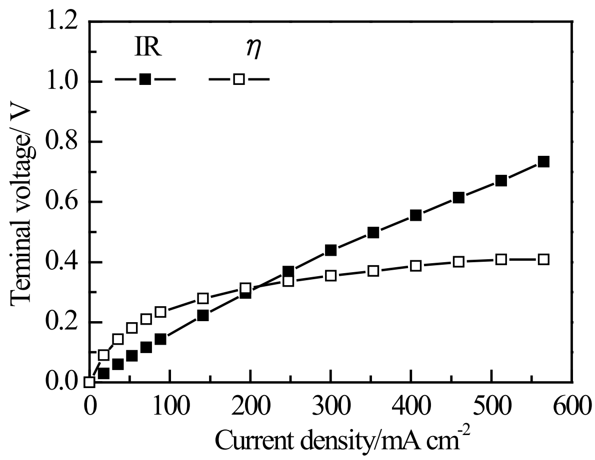

Fig. Ohmic loss, *IR*, (filled square) and electrode overpotential, *η*, (open square) of steam electrolysis (600°C). Anode: $Sm_{0.5}Sr_{0.5}CoO_3$ (SSC55); electrolyte: $SrZr_{0.5}Ce_{0.4}Y_{0.1}O_{3-\alpha}$ (SZCY541); cathode: cermet of porous nickel and SZCY541.

[1] T. Sakai, S. Matsushita, H. Matsumoto, S. Okada, S. Hashimoto and T. Ishihara, "Intermediate temperature steam electrolysis using strontium zirconate-based protonic conductors", International Journal of Hydrogen Energy, **34**, 56 (2009).