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

Matsumoto, H. (IFRC, Kyushu Univ.)

Introduction

Assuming the use for hydrogen isotope sensing, hydrogen pumps and steam electrolyzers using "proton-conducting oxides" has been investigated. Proton conducting oxides typically belong to ABO_3 perovskites to which aliovalent cation doping is conducted. The oxides operate even at 600°C and are thus useful as electrolytes for electrochemical devices working at around the temperature. 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]. So far, a thin film of SZCY541 fabricated on the substrate of NiO/SZCY541composite was found to work with reduced ohmic resistance and hence high energy efficiency [2]. In this study, the effect of volumetric fraction of NiO/SZCY541 in the composite on the steam electrolysis properties was examined.

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 [1]. The obtained SZCY541 powder was mixed with NiO powder with volume fraction from 4/6 to 6/4, and sintered at 1400°C. The electrical conductivity of the composite specimen reduced in hydrogen (resulting in Ni/SZCY541) was measured. The composite in pellet form with 14 mm diameter and 0.5 mm thickness (Fig. 1a) was used as a cathode substrate. SZCY541 paste composed of the powder, ethyl cellulose and a solvent was screen printed on to the NiO/SZCY541 cathode substrate (Fig. 1b) and co-fired at 1400°C to prepare thin SZCY541 layer on the substrate (Fig. 1c). 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; Fig. 1d). 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 Fig. 2). 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 generation was evaluated by gas chromatography and overvoltage character was measured by a current interrupt method, respectively.

Results and Discussion

Ni/SZCY541composite with a volume fraction of 4/6 showed low electronic conductivity, suggesting insufficient volume or Ni causing poor network formation. Those with

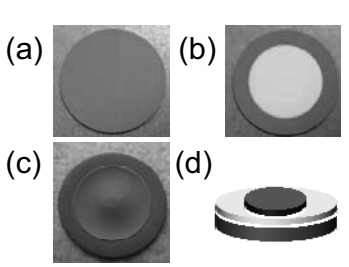

Fig. 1 preparation of specimen

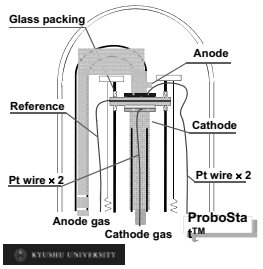

Fig. 2 Electrolysis cell

fractions of 5/5 and 6/4 had enough conductivity and were used as the substrate of electrolysis cells. The result of electrolysis experiments is shown in Fig. 3. Electrode overpotential of the cell with Ni/SZCY=6/4 is lower than that with 5/5, suggesting larger amount of reaction cites at the Ni/SZCY interface. In contrary, the cell with Ni/SZCY=6/4 had a higher Ohmic loss than the other, probably due to the higher resistivity of the cathode substrate. The latter factor, the Ohmic losses, has larger effect the total *i-V* characteristics, and as a result, the cell with Ni/SZCY=5/5 showed the best performance. On the basis of the above discussion, a tubular cell is being constructed with jigs shown in Fig. 4.

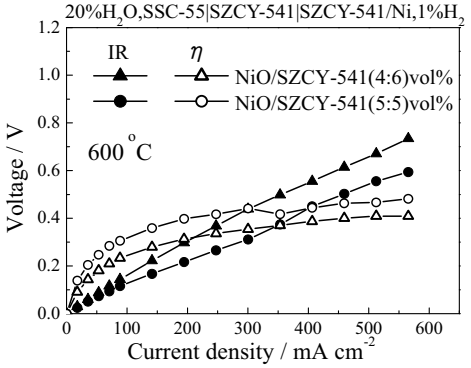

Fig. 3 Voltage characteristics of steam electrolysis performed at 600°C: Ohmic loss (closed markers) and electrode overpotentials (open makers) for the fraction of NiO to electrolyte 4/6 and 5/5 by volume.

Fig. 4 Jigs for the preparation of tubular cells

[1] T. Sakai, et al., Int. J. Hydrogen Energy, **34**, 56 (2009)
[2] H. Matsumoto, Annual Report of NIFS, pp. 328 (2011)