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

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Introduction

Proton conducting oxide is typically ABO_3 perovskite, receiving partial atomic substitution for either A or B site, and work as an electrolyte operating at 600°C. Since the charge carrier is proton, electrochemical pumping of hydrogen isotopes including tritium is possible and thus applicable to the hydrogen isotope detector. In this study, the use of the proton conducting oxide for hydrogen isotope separation. We have investigated so far the hydrogen pumping and steam electrolysis, and elucidated that the hydrogen pumping is possible with relatively high current efficiency, and that water molecule introduced to the anode compartment can be split to hydrogen and oxygen electrochemically [1, 2].

Structure of Electrochemical cell used for the experiment of steam electrolysis is shown in Fig. 1. Anode such as $Sm_{0.5}Sr_{0.5}CoO_3$ contains transition metals, and Ni is used as the cathode. The transition metals possibly deteriorates the performance of the cell, during processing and operation. Therefore in this study, electrochemical properties of $SrZr_{0.9}Y_{0.1}O_{3-\alpha}$ (10Y-SZO) on the introduction of Fe, Co, Mn was examined.



Fig. 1 Steam electrolysis and the structure of anode/electrolyte/cathode.

Experiment

SrCO₃, ZrO₂, Y₂O₃ and transition metal oxides were used as starting materials. The appropriately weighed amounts were mixed and fired in air at 1300°C, followed by ball milling, drying and finally fired in air at 1600°C to obtain dense ceramic samples. Bar-shape electrolyte is used for the conductivity measurements and disk-shape electrolyte is used for the gas concentration measurements.

Results and Discussion

Fig. 2 (upper) shows the conductivity of 10Y-SZO containing Co, Ni and Fe at 1mol% measured in moist hydrogen. It is clearly seen that the introduction of the transition metals causes the decrease in the conductivity. The same tendency was observed for the conductivity measured in moist air. The change of the conductivity is shown as a function of introduced dopant in Fig. 2 (lower). Particularly, the introduction of Fe leads to significant reduction of 1% Fe. The introduction of Co and Ni also cause the decrease in the conductivity. On the other hand, the electromotive force of hydrogen concentration cells agree with that calculated from Nernst equation for all transition-metal-introduced 10Y-SZO.

From these results, it is conclusive that the introduction of the transition metals decrease the conductivity and this tendency is much significant in the case of Fe. This suggests that the choice of the electrode materials should be made not only from the aspect of electrode performance, but also from the impact on the electrolyte conductivity.



Fig. 2 electrical conductivity of $\text{SrZr}_{0.89}\text{Y}_{0.1}M_{0.01}\text{O}_{3-\alpha}$ (*M*=Co, Ni, Fe) in moist hydrogen (upper) and dependence of the conductivity on the transition metal dopants (lower).

Sakai, T., et al., Int. J. Hydrogen Energy, 34, 56 (2009).
Matsumoto, H., et al., Pure Appl. Chem., 85,427 (2013).