

§2. Hydrogen Isotope Separation Using Proton-conducting Oxides

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Proton conducting oxide uses ABO_3 perovskite as a mother structure, receiving partial atomic substitution for either A or B site, and is suitable electrolyte for operation of electrochemical cells at intermediate temperatures, such as 600°C. Since the charge carrier is proton (hydrogen ion), electrochemical pumping of tritium is possible. In this study, we aim to use this material of proton conducting oxide for hydrogen isotope separation. We have investigated so far the hydrogen pumping and steam electrolysis which serve for isotope separation, and elucidated that the hydrogen pumping is possible with relatively high current efficiency, and that water molecules introduced in the form of steam to the anode compartment can be split to hydrogen and oxygen electrochemically [1].

In the case of steam electrolysis, $SrZr_{0.9}Y_{0.1}O_{3-\alpha}$ and $SrZr_{0.5}Ce_{0.4}Y_{0.1}O_{3-\alpha}$, and etc. are typically used as the electrolyte. Nickel and transition-metal-containing metal oxide such as $Sm_{0.5}Sr_{0.5}CoO_3$ are used as the cathode and anode, respectively. These electrodes are attached to the electrolyte by backing at high temperature. During this process, as well as in operation of electrolysis, electrode and electrolyte materials possibly react with each other, resulting in deterioration of the electrolyzer. Therefore, the effect of introducing transition metals into the perovskites on the electrical properties has been investigated in this study. The effect arising from 1%-transition-metal introduction has been reported in the past. The results of 5%-transition metal introduced to the perovskite with Ba or Sr at the A -site and Ce or Zr at the B -site, respectively.

$AB_{0.85}Y_{0.1}M_{0.05}O_{3-\alpha}$ ($A=Ba, Sr$; $B=Ce, Zr$; $M=Co, Ni, Fe, Mn$) was prepared by a solid state reaction method reported elsewhere [1]. The specimens were bar-shape and the conductivity was measure in wet hydrogen and oxygen.

$SrZr_{0.9}Y_{0.1}O_{3-\alpha}$ (SZY91), $SrCe_{0.9}Y_{0.1}O_{3-\alpha}$ (SCY91), and $BaCe_{0.9}Y_{0.1}O_{3-\alpha}$ (BCY91) were used as the base proton conducting oxides. The conductivities of the materials introducing 5% transition metals are shown in Figs. 1 (a)-(c). The measurements shown in the figures were conducted in moist air. In the case of BCY91, the results are consistent with a previous report and conductivity is seen to decrease on the introduction of transition metals, particularly in the case of manganese resulting in the drop of the conductivity larger than one decade. This behavior is very similarly observed in the case of SZY91, i.e., the conductivity decreases with introducing transition metals to SZY91 system. On the other hand, the conductivity slightly increases in the case of SCY91 for the introduction of transition metals. These results reveals that transition metals are toxic to the proton conducting perovskite in many cases, and that there is an exceptional case, SCY91 in this study,

for the decreasing conductivity. It is experimentally evident that the increase and decrease of the conductivity depends on the choice of both proton-conducting base oxide and transition metals and that the combination of the electrolyte and electrode should be optimum to get the high performance of hydrogen isotope separation.

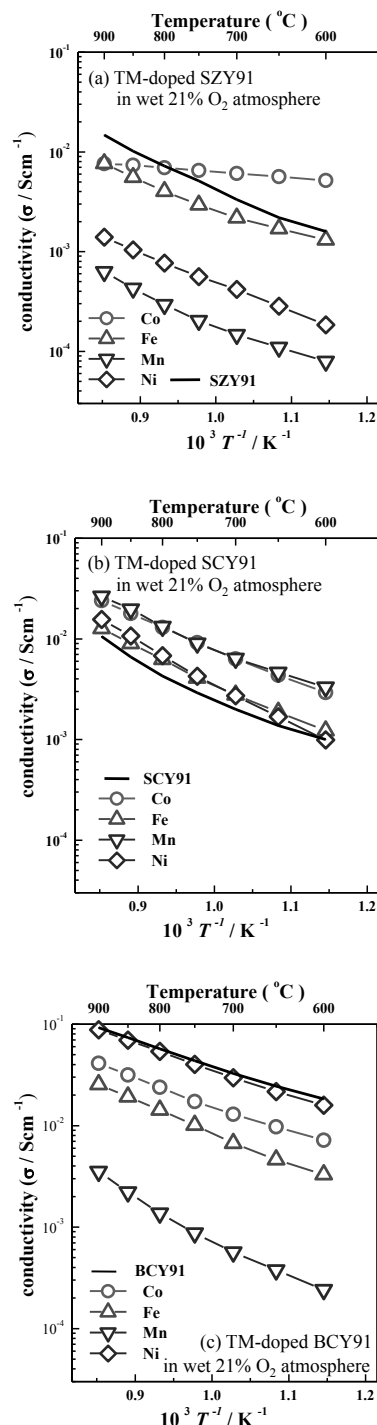


Figure 1 Conductivity of transition-metal-doped perovskites measure in wet 21% O₂: (a) SZY91, (b) SCY91 and (c) BCY91 system.

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