§8. Hydrogen Separation and Sensing Using Proton-Conducting Oxides

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Nuclear fusion reactors based on the use of hydrogen isotopes needs efficient hydrogen separation and sensing. This study deals with the material designing and examination of the proton-conducting oxides. We intend the use of the materials for electrochemical hydrogen pumps, which enables us hydrogen separation with high energy efficiency and selectivity, and hydrogen sensing with high sensitivity.

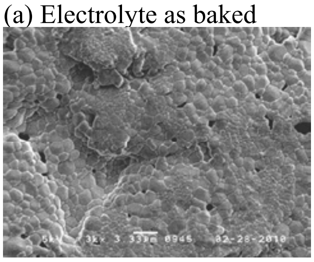
Typical proton-conducting oxides have a Perovskite-type crystal structure with a chemical formula of ABO_3 (typically, *A*=Ca, Sr, Ba; *B*=Ce, Zr）. Aliovalent doping to the *B* site leads to the formation of protonic defects that act as an ion-conducting charge carrier. $CaZr_{0.9}In_{0.1}O_{3-\alpha}$ (below denoted as CZI91) is one of the most suitable proto-conductor due to its chemical stability. We previously found that palladium electrode has good electrocatalytic activity when used with $SrZrO_3$-based proton-conducting oxides [1]. It is assumable that the combination of the $CaZrO_3$-based proton conductor and the palladium electrode will be suitable for the hydrogen isotope separation and sensing in the nuclear fusion reactor; we can expect both the performance and reliability for the combination of the CZI91 electrolyte and Pd electrode. This year, preparation of the palladium electrode on to the CZI91 was examined.

Disk-shape samples of CZI91 (13 mm diameter and 1.6 mm thickness) were purchased from TYK Corporation, prepared via solid state reaction route. Palladium paste (Tanaka Kikinzoku Kogyo, T-80) was screen printed to either polished or unpolished surface of the CZI91 disks; polishing was finished with abrasive paper of #500 fine. The specimen was dried and baked in air for 2h. Three baking temperatures were examined: 800, 900 or 1300°C.

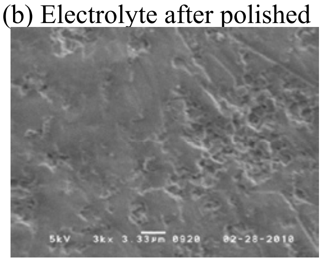
SEM pictures of the CZI91 electrolyte samples and palladium electrodes baked on to the surface of the electrolyte are shown in the figures below. As shown in Figures (a) and (b), the surface of the as prepared CZI91 was made of grains in the dimension of several microns and the polishing removed the texture of the grains from the surface. The microstructures and morphology of the palladium electrode differ largely depending on the baking temperature. Comparing figures (e) and (f), baking at 800°C resulted in the formation of porous structure, and that at 1300°C essentially makes the electrode fully dense; densification occurred even at 900°C, and thus the critical temperature may lie between 800-900°C for the densification. When we look at the edge of the deposited electrode, densification occurred after baking at 1300°C. Polishing of the surface has no significant effect on the state of palladium electrode.

These results well agreed with those we previously obtained for $SrZr_{0.9}Y_{0.1}O_{3-\alpha}$[1]. In the case of electrochemical hydrogen pumps using $SrZr_{0.9}Y_{0.1}O_{3-\alpha}$, electrode activity became much higher for the dense palladium electrode (like the structure in figure (f)) that for the porous electrode (figure (e)). Therefore, high electrode performance will be expected for the dense palladium electrode, prepared by baking at 1300°C and further investigation will be continued.

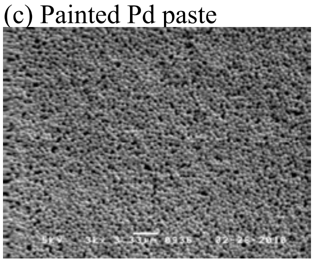
(a) Electrolyte as baked



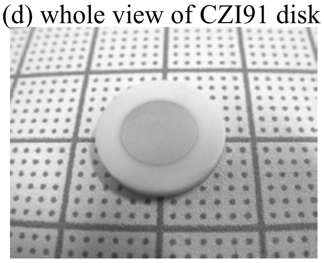
(b) Electrolyte after polished



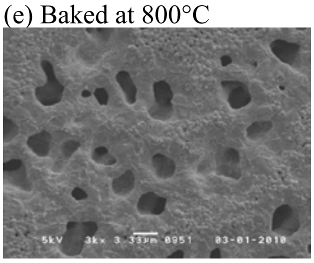
(c) Painted Pd paste



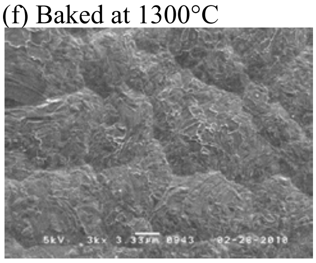
(d) whole view of CZI91 disk



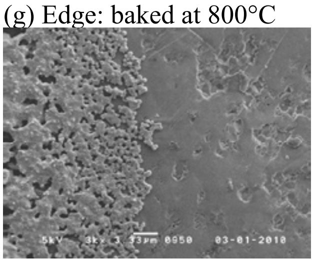
(e) Baked at 800°C



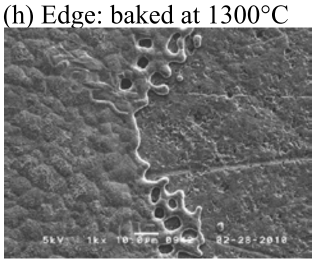
(f) Baked at 1300°C



(g) Edge: baked at 800°C



(h) Edge: baked at 1300°C



Figures Scanning electron microscope images of CZI91 electrolyte and palladium electrodes

1) Sakai, T. et al., Ionics, 15 (2009) 665-670.