Development of new cathode materials and infiltration method for Intermediate-Temperature Solid Oxide Fuel CellsExample of Pr_6O_{11}

Jean-MarcBASSAT(1), ClémentNICOLLET(1), Vaibhav VIBHU(1), Aurélien FLURA(1), Aline ROUGIER(1),Jean-Claude GRENIER(1)

1CNRS, Université de Bordeaux, ICMCB,87 Av. Dr Schweitzer, Pessac Cedex, F-33608, France
jean-marc.bassat@icmcb.cnrs.fr

The present study is focused on alternative oxygen electrodes for Intermediate Temperature Solid Oxide Fuel Cells (IT-SOFCs, 600<T°C<700) especially using Metal Supported Cells (MSCs). For the latter, to prevent detrimental oxidation of the metal support, sintering of the cell components at high temperature under low pO_2 is required. $Ln_2NiO_{4+\delta}$ (Ln=La, Pr, Nd) compounds with the K_2NiF_4 type structure act as alternative cathode materials for IT-SOFC due to their mixed ionic and electronic conductivity. $Pr_2NiO_{4+\delta}$ shows excellent electrochemical properties at intermediate temperature (*i.e.* low polarization resistance R_p value, R_p = 0.03 Ω.cm^2 at 700 °C), while $La_2NiO_{4+\delta}$ exhibits higher chemical stability. Thus, the properties of $La_{2-x}Pr_xNiO_{4+\delta}$ mixed nickelates were investigated with the aim to find best compromise between chemical stability and electrochemical performances. Herein, the chemical stability of the nickelates under air at operating temperatures as well as the evolution of the polarization resistances during ageing (recorded under air at i_{dc} = 0 and $i_{dc} \neq 0$ conditions) were studied for duration up to one month. $La_2NiO_{4+\delta}$ is chemically stable whereas $Pr_2NiO_{4+\delta}$ dissociates after 1 month at 600, 700 and 800 °C, with formation of, in particular, *Pr_6O_{11} oxide*.

One way to increase the cathode electro catalytic properties and decrease its polarization resistance is to increase the Triple Phase Boundaries zone, where the oxygen is reduced. A promising solution to achieve this goal is to disperse an oxygen reduction catalyst on the surface of a porous ionic conductor. In this study, cathodes of lanthanum nickelate *infiltrated* into Gd-doped ceria backbone were prepared. Optimization of the preparation parameters led to a large decrease of the polarization resistance, down to 0.1 Ω·cm^2 at 600 °C. The influence of the preparation parameters of the composite electrodes on their electrochemical activity will be presented. The cathodes were then applied onto a commercial anode-electrolyte half-cell and complete cell polarization measurements showed power density higher than 1 W.cm^{-2} at 800°C.

The praseodymium oxide, Pr_6O_{11}, is regarded as a potential electro catalyst for the oxygen reduction reaction. First the ionic conductivity and oxygen surface exchange coefficient were measured by isotopic exchange depth profiling combined with secondary ion mass spectroscopy. At 600 °C, the value of the oxygen diffusion coefficient is as high as 3.4x10^{-8} cm^2.s^{-1}, and that of the surface exchange coefficient is 5.4x10^{-7} cm.s^{-1}, which supposes excellent electro catalytic properties. The measured electronic conductivity is high enough for using this material as a SOFC cathode. Herein, praseodymium nitrate was *infiltrated* into Gd doped ceria (GDC) backbone and fired at 600 °C to form a composite oxygen electrode Pr_6O_{11}/GDC. Electrochemical measurements show very low polarization resistance, *R*p = 0.028 Ω cm^2 at 600 °C, which is among the best values ever reported for SOFC cathodes. Then, a single cell made of a commercial Ni-YSZ/YSZ half-cell and of the infiltrated cathode is able to deliver a maximum power density of 825 mW cm^{-2} at 600 °C. Finally, ageing of this cell for 840 h, at 600°C and 0.5 A cm^{-2}, shows a degradation rate lower than 1%.