

Validation of $\text{Nd}_2\text{NiO}_{4+\delta}$ as Oxygen Electrode Materials for Intermediate Temperature Solid Oxide Cells with LSGM Electrolytes.

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The search of new electrode materials is an active field of research. Particularly, oxygen electrode materials for Solid Oxide Cells (SOC) must fulfill several requirements such as high electrocatalytic activity for oxygen redox reaction, and good thermal and chemical compatibility with solid electrolytes.

The $\text{Ln}_2\text{NiO}_{4+\delta}$ (Ln = lanthanide) compounds belong to the Ruddlesden-Popper series and have the aptitude to accommodate oxygen ions into interstitial sites in the LnOx layer. These ions present high mobility giving place to high ionic conductivity by interstitial oxygen diffusion [1]. Therefore, these nikelates could be oxidized under anodic polarization, producing Ni^{+3} ions and oxygen interstitials defects, which increase the electrocatalytic properties and make these compounds suitable to be operated in Electrolyzes mode [2]. These materials $\text{Ln}_2\text{NiO}_{4+\delta}$ also present Thermal Expansion Coefficients (TEC) similar to the most widely used SOFC or IT-SOFC electrolytes: $\text{Zr}_{0.92}\text{Y}_{0.08}\text{O}_{1.96}$ (YSZ), $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{1.96}$ (CGO) and $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{2.85}$ (LSGM) [3-5]. However, one drawback of these materials is the chemical reactivity with CGO and YSZ electrolytes at operation (600 to 800 °C) or adhesion temperatures (900 - 1000 °C) [6-8]

In this work is presented the study of electrode mechanism using Electrochemical Impedance Spectroscopy (EIS) for $\text{Nd}_2\text{NiO}_{4+\delta}/\text{LSGM}/\text{Nd}_2\text{NiO}_{4+\delta}$ symmetrical cells. The temperature was changed between 600 and 800 °C while the oxygen partial pressure pO_2 varies between 0.2 and 10^{-4} atm. No decomposition or chemical reaction between LSGM and $\text{Nd}_2\text{NiO}_{4+\delta}$ (NNO) was found. The influence of the microstructure of the cathodes was considered, using NNO prepared by three different synthesized methods, two chemical routes (HMTA and CIT) and conventional solid state reaction (SSR). Figure 1a show as the Area Specific Resistance (ASR) decrease as the particle size decrease and the surface area increases. In Figure 1b, the ASR values are compared with previous study on NNO/CGO/NNO cells [9].

From the study of dependence of ASR with pO_2 three contributions were identified: at high, medium and low frequencies. The high frequencies contribution present in all NNO/LSGM systems was associated to the oxygen ion transference from the electrode to the electrolyte. At difference to those found for NNO/CGO half cells, the medium frequencies contribution was attributed to interstitial oxygen diffusion in the solid. In NNO/CGO case, the reactivity between electrode and electrolyte diminished the active zone for oxygen reaction introducing a co-limiting process, the oxygen surface exchange, to the oxygen diffusion [9]. The low frequencies contribution was related to dissociative adsorption in samples obtained by HMTA and CIT, whereas in SSR sample where the particle size is higher

and the porosity is lower the gas diffusion process become important and increases the polarization resistances.

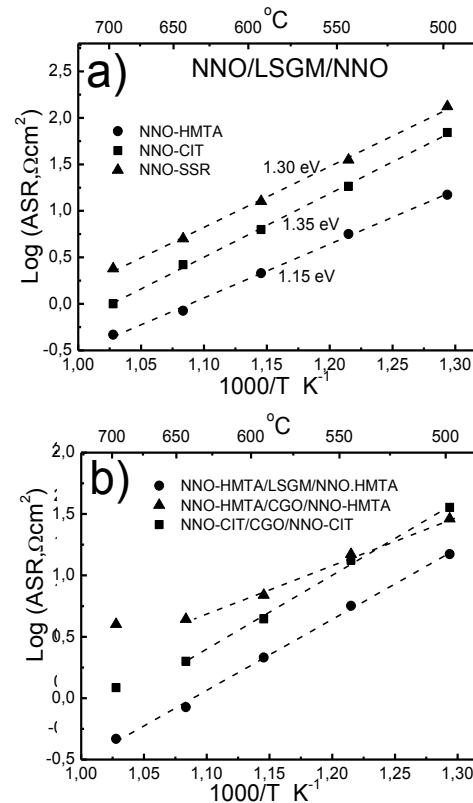


Figure 1. a) Dependence of the ASR with temperature for NNO-HMTA, NNO-CIT and NNO-SSR electrodes deposited on LSGM electrolyte. b) Comparison of ASR values for NNO-HMTA/LSGM/NNO-HMTA cell against NNO-HMTA/CGO/NNO-HMTA and NNO-HMTA/CGO/NNO-HMTA cells [14].

References

- [1]. F. Mauvy, J.-M. Bassat, E. Boehm, J.-P. Manaud, P. Dordor, J.-C. Grenier. Solid State Ionics 158 (2003) 17–28.
- [2] Chauveau, F., Mougin, J., Bassat, J.M., Mauvy, F., Grenier, J.C. Journal of Power Sources 2010, 195 (3) , 744-749.
- [3]. Kharton VV, Viskup AP, Kovalevsky AV, Naumovich E, Marques FMB. Solid State Ionics 2001; 143:337–53.
- [4]. Minervini L, Grimes RW, Kilner JA, Sickafus KE. Journal of Materials Chemistry 2000; 10:2349–54.
- [5]. Skinner SJ, Kilner JA. Solid State Ionics 2000;135:709.
- [6]. Bassat J.M., Odier P., Villesuzanne A, Marin C, Pouchard M. Solid State Ionics 2004; 167:341–7.
- [7] Montenegro-Hernández A, Mogni L, Caneiro A. International Journal of Hydrogen Energy 35(11) (2010) 6031-6036
- [8] Montenegro-Hernández A, Vega-Castillo JE, Mogni L, Caneiro A. International Journal of Hydrogen Energy 36(24) (2011) 15704-15714.
- [9] Montenegro-Hernández A, Mogni L, Caneiro A. International Journal of Hydrogen Energy 37(23) (2012) 18290-18301.