ELECTROCHEMICAL BEHAVIOUR OF Ni-$Ce_{0.9}Gd_{0.1}O_{2-\delta}$ SOFC ANODES IN METHANE

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Solid Oxide Fuel Cells (SOFC) exhibit a significant advantage with the ability of operation on natural gas. The direct utilisation of natural gas makes SOFC, amongst the other fuel cell systems, potentially more competitive within the present technologies for energy conversion. Methane, the main component of natural gas, can be directly converted into a mixture of hydrogen, water, carbon monoxide and carbon dioxide by steam reforming on an SOFC anode. The state-of-the-art SOFC anode is a Ni-YSZ cermet. Ni has a high catalytic activity for the dehydrogenation of hydrocarbons. Ni-YSZ cermet anodes have the tendency to crack methane which results in a carbon deposition with high risk for cell deactivation. Consequently, a steam/methane ratio higher than 2 is required to avoid carbon deposition.

Recently, there are increasing efforts in the development of SOFC anodes capable of operation in natural gas, without suffering from carbon build up due to catalytic cracking. In this regard, ceria-based materials are of potential interest. Such ceria-based anodes have important advantages over conventional Ni/YSZ anodes, namely the ability to suppress carbon deposition on electrodes in methane-rich atmospheres. Therefore, the cell can operate on methane with low steam/methane ratios. Furthermore, the high concentration of oxygen vacancies and the mobility of lattice oxygen in the mixed conductor doped ceria makes it possible to extend the triple phase boundary (TPB) from the anode/electrolyte interface onto the whole surface of the anode. However, the electronic conductivity of doped ceria is not sufficient to take care of the current collection in an SOFC stack, and an electronic contribution with high conductivity in the anode is needed in order to prevent lateral electrical losses. Furthermore, ceria has a low activity for methane oxidation. Introduction of Ni into the doped ceria matrix is a way to acquire sufficient conductivity and break the C-H bond more easily.

In this study, the electrochemical performance of Ni-$Ce_{0.9}Gd_{0.1}O_{2-\delta}$ (Ni-CGO) with NiO contents of 2.5 and 70 %wt were investigated at high temperatures in 10%CH_4/90%N_2 and CH_4 which was humidified at ambient temperature. Stability tests were carried out in methane humidified at ambient temperature to characterize the tolerance of the anode with 70 wt% NiO content to carbon deposition. The microstructure of the electrodes before and after the electrochemical investigations and stability tests were examined by SEM. The anode with 70 wt% NiO was investigated by impedance spectroscopy at high steam/methane ratios as well.

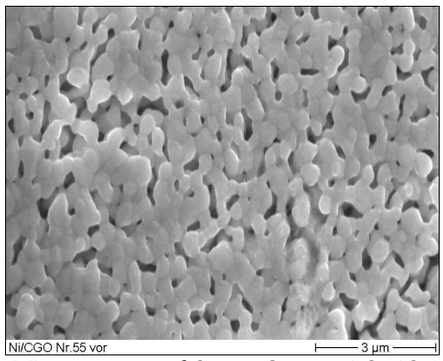

Fig. 1: *Microstructure of the anode material with 2.5 wt% NiO before the electrochemical measurements.*

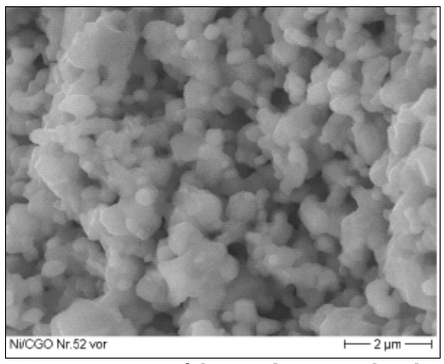

Fig. 2: *Microstructure of the anode material with 70 wt% NiO before the electrochemical measurements.*

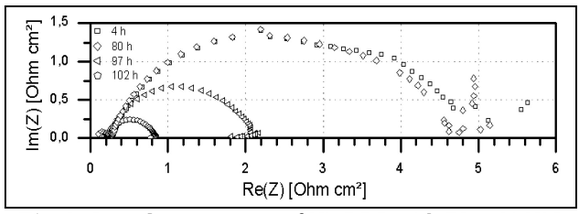

Fig. 3: *Impedance spectra of Ni-CGO with 70 wt% NiO in 97% CH_4/3% H_2O at 850 °C during the aging process.*

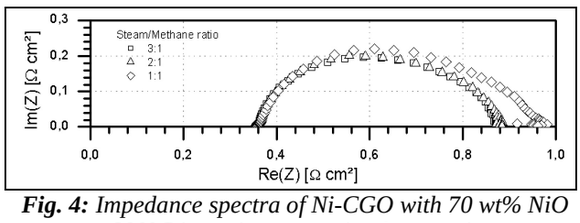

Fig. 4: *Impedance spectra of Ni-CGO with 70 wt% NiO at 800°C in various steam/methane ratios.*

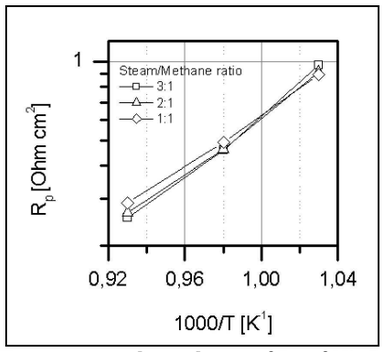

Fig. 5: *Temperature dependence of R_p of Ni-CGO with 70 wt% NiO in various steam/methane ratios.*