

Operation of barium zirconate-supported planar solid oxide fuel cells at 600°C with methane

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Abstract – Unitary planar electrochemical cells of the type anode (Pt film)/solid electrolyte (barium zirconate or barium cerate protonic conductor)/cathode (Pt or La_{0.6}Ca_{0.4}Fe_{0.8}Co_{0.2}O_{3-δ}-LCFC film) were assembled for evaluation of open circuit voltage (OCV) under methane and hydrogen in the 300-600°C temperature range. BaZr_{1-x}Y_xO_{3-δ}, x=0.08, 0.15 and 0.2, BaCe_{0.8}Gd_{0.2}O_{3-δ} and LCFC were prepared by solid state synthesis. A niobium phosphate based glass was used as sealant. The cells operating with methane being injected to the anode showed OCV values similar to the ones measured in cells with gadolinium-doped barium cerate (BaCe_{0.8}Gd_{0.2}O_{3-δ}) solid electrolytes, prepared for comparison purposes for both (Pt/solid electrolyte/Pt) and (Pt/solid electrolyte/LCFC) configurations. However, under operation with 4%H₂+96%N₂, the barium cerate cells presented higher values of OCV than the barium zirconate cells. The reaction of methane at the Pt-proton conductor triple boundary phase was found to depend strongly on the fuel flux, the same not being true for hydrogen. The optimum temperature for methane reacting with platinum for proton production was in the 500-520° range.

Barium zirconate, BaZrO₃, is a well known refractory material with very high melting point (2600 °C), small thermal expansion coefficient, poor thermal conductivity, good mechanical properties, thermal stability and low chemical reactivity towards corrosive compounds [1].

In this work the following barium zirconate compositions BaZr_{0.92}Y_{0.08}O_{3-δ}, BaZr_{0.8}Y_{0.2}O_{3-δ} and BaZr_{0.85}Y_{0.15}O_{3-δ} were prepared by solid state synthesis. The powders were attrition milled for increasing the final sintered density. The powders were analyzed by X-ray diffraction (crystalline phases) and laser scattering (distribution of particle size). Simultaneous thermogravimetric and differential thermal analyses were performed for the study of the thermal behavior during the formation of the compounds. Pressed powders were sintered and characterized by determining a) apparent density by the Archimedes method with kerosene as liquid medium, b) crystalline phases by X-ray diffraction (Figure 1), c) morphology by scanning electron microscopy of fracture surfaces. Densification was best achieved for BaZr_{0.92}Y_{0.08}O_{3-δ} and reached 96% of theoretical density upon sintering at 1500°C. Unitary planar electrochemical cells of the type anode (Pt)/protonic conductor/cathode (Pt or La_{0.6}Ca_{0.4}Fe_{0.8}Co_{0.2}O_{3-δ}) were assembled for evaluation of open circuit voltage (OCV) in the 300-600°C range, and current density at 600°C, both under methane (Figure 2) and hydrogen.

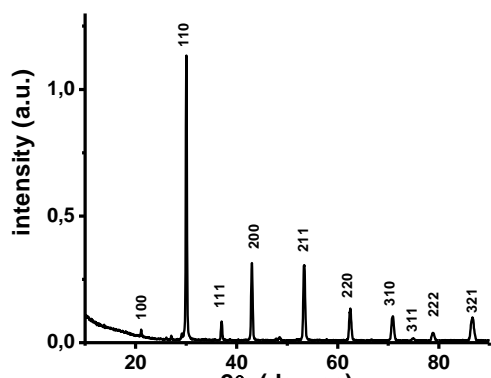


Figure 1: X-ray diffraction pattern of BaZrYO pellets sintered at 1500°C.

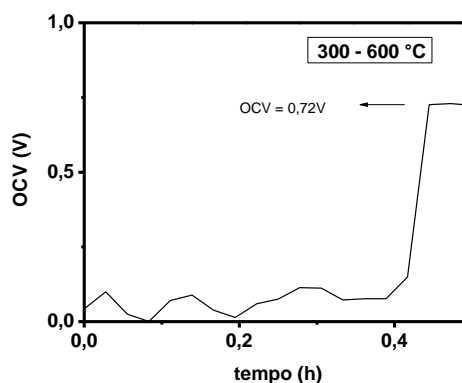


Figure 2: Open circuit potential of LCFC/BaZr_{0.85}Y_{0.15}O_{3-δ}/Pt single solid oxide fuel cell under methane in the 300-600 °C range.

Reference

[1] A. M. Azad, S. Subramaniam, T. W. Dung, "On the development of high density barium metazirconate (BaZrO₃) ceramics", *Journal of Alloys and Compounds* **334** (2002) 118.