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Correlation between microstructure and thermodynamic stability of Y^{+3}-doped $BaCeO_3$ in water vapor and CO_2-containing atmosphere

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Abstract – This work presents the correlation between powders characteristics, synthesized by solid-state reaction and citrate process routes, and stability of $BaCe_{0,9}Y_{0,1}O_{3-\delta}$ electrical conductivity.

Barium cerate ($BaCeO_3$) doped with trivalent ions contains oxygen vacancy due to the substitution of Ce^{+4}. Besides the oxygen vacancy, $BaCe_{1-x}M_xO_{3-\delta}$ becomes a protonic (H^+) conductor when heat treated under hydrogen or humid atmosphere. Due to the high protonic conduction, doped barium cerate ceramics are known to be potentially useful as solid electrolytes in high temperature fuel cells and hydrogen gas sensors. However, despite the promising characteristics, the literature shows that this material is unstable under high temperature in CO_2-containing atmosphere or water vapor [1]. The non stability is described by reactions (1) and (2),

$$BaCeO_3(s) + CO_2(g) \rightarrow BaCO_3(s) + CeYO_2(s) \quad (1)$$

$$BaCeO_3(s) + H_2O(g) \rightarrow Ba(OH)(s/l) + CeYO_2(s) \quad (2)$$

In this work the $BaCe_{0,9}Y_{0,1}O_{3-\delta}$ powders were synthesized by solid-state reaction and citrate process routes. The samples were isostatic pressed and sintered following two sintering routes. Samples prepared with microsized powder were sintered with the conventional method while the two step sintering process, figure 1, was used for nanosized powder.

The powder prepared by citrate process was characterized by transmission electron microscopy (TEM). The particles are nanometric as shown the figure 2.

The main techniques used for sintered samples characterization were X-ray diffraction, scanning electron microscopy and impedance spectroscopy in H_2O-containing atmosphere

Figure 3 shows the Y-$BaCeO_3$ X-ray diffraction patterns obtained from microsized powders pellets sintered at 1600 °C with 4, 8, and 12 hours of dwelling time. As predicted by equation (1), it was found $CeYO_2$ beyond the phase of interest. The nanosized powder pellets sintered through two step process are dense and the grains are nano scaled.

The impedance spectroscopy analysis was sensitive to monitor the non stability of the powders as well as the sintered samples heat treated in different atmospheres.

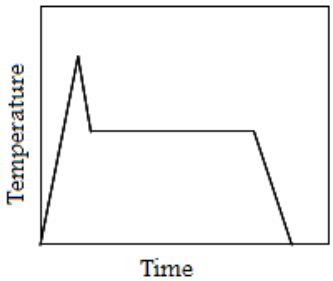

Figure 1: Two step sintering

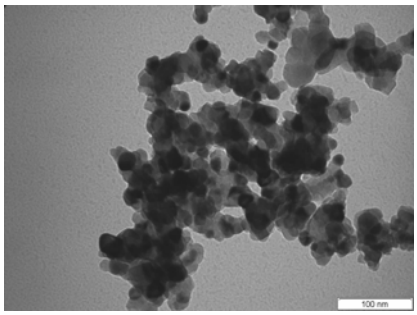

Figure 2: TEM image of a Y-$BaCeO_3$ particle synthesized by citrate process routes, calcined at 700°C/2h.

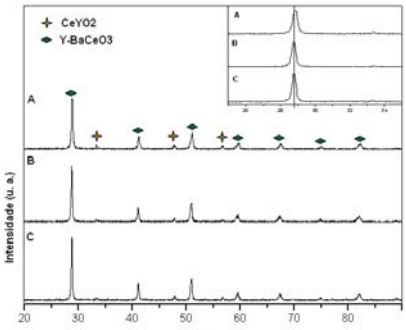

Figure 3: Y-$BaCeO_3$ X-ray diffraction patterns. Dwelling **a)** 12 hours; **b)** 8 hours; **c)** 4 hours

[1] TANNER C. W.; VIRKAR V.; Journal Electrochemical Society; v. 143; p. 1386, 1996