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Impedance spectroscopy of YSZ electrolyte containing CuO for various applications

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Abstract

The electrical properties of yttria stabilized zirconia (YSZ) electrolyte containing different amounts of CuO (0–1 mol%) have been studied. The complex impedance diagrams showed an increase in the grain boundary resistivity with CuO addition. The bulk resistivities were comparable to those of the CuO free YSZ except for the sample containing 1 mol% CuO which showed a small increase in resistivity. The results showed the possibility to sinter YSZ at low temperature 1250 °C to a very high sintered density by adding a small amount of CuO (0.3 mol%). The conductivity of the latter is nearly equal to that of YSZ sintered at 1500 °C. This might be a step forward in the development of oxygen pumps by reducing the electrolyte sintering temperature which leads to better quality and low cost.

Keywords: Impedance spectroscopy; Electrical properties; Electrolyte; 8YSZ

1. Introduction

Yttria stabilized zirconia is a very interesting material in many applications such as: the solid electrolyte in oxygen sensors [1], oxygen pumps [2] and in solid oxide fuel cells (SOFCs) due to its high ionic conductivity. In addition it has a good mechanical stability and can be used over a wide temperature range. In a gasoline engine the control of air to fuel ratio could positively contribute to pollutant emission reduction. This could be achieved by using oxygen sensors made of YSZ electrolyte layer deposited on a porous alumina substrate [3]. The firing process of the electrolyte layer/substrate composite is a critical one due to the differential shrinkage during heating up and the thermal expansion mismatch during the cooling down. This problem could be reduced if the electrolyte layer could be sintered at a relatively low temperature.

The reduction of the firing temperature of the electrolyte layer contributes to the reduction of the fabrication cost. However, we have to make sure that the performance – in this case – remains at the same level as when the electrolyte was fired at high temperature. Various transition metal oxides were added to yttria stabilized zirconia (YSZ) [4–6]. These additions were intended to promote sintering and grain growth. However, the additions in large quantities were found to induce electronic conduction. In a previous work [7] it was found that the addition of CuO enhances the sinterability of yttria stabilized zirconia. The aim of this work is to study the effect of the addition of CuO in small amounts on the electrical properties of the yttria stabilized zirconia membrane.

2. Experimental procedures

A Tosoh fully stabilized powder of 8 mol% Y_2O_3–ZrO_2 having a BET surface area of 13 m^2/g and crystallite size of

Table 1
Density of 8YSZ+*x* CuO, sintered at 1250 °C and 1500 °C for 1 h

Sintering temp.	1250 °C		1500 °C	
Sample	Density, g/cm^3	% of the T.D. [a]	Density, g/cm^3	% of the T.D.[a]
Pure 8YSZ	4.575	75	5.868	96
8YSZ+0.3 mol% CuO	5.782	95	5.886	97
8YSZ+0.5 mol% CuO	5.667	93	5.865	96
8YSZ+1.0 mol% CuO	5.307	87	5.793	95

[a] Theoretical density (T.D.) taken as 6.1 g/cm^3 neglecting the effect of small amounts of CuO remaining after sintering ±0.3%.

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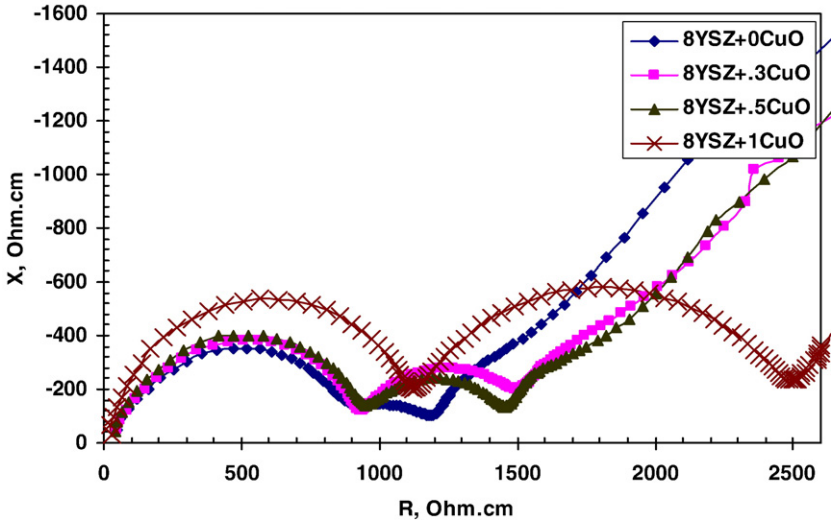

Fig. 1. Impedance spectroscopy for 8YSZ+xCuO sintered at 1500 °C and tested at 500 °C.

26 nm as given by the supplier was used in this work. The powder in the as received form was in agglomerated spherical shape. This powder was used to prepare samples containing 0, 0.3, 0.5 and 1 mol% CuO. The appropriate weights of $CuSO_4$ were dissolved in distilled water and then mechanically mixed with 8YSZ. The material was dried then calcined at 700 °C for 1 h. The calcined powder was milled in a ball mill with ethyl alcohol using tetragonal zirconia balls for 24 h. The milled powders were dried at 120 °C for 24 h then crushed and sieved.

The powders pressed in a 5 mm diameter and 2 mm thickness disk shape were sintered in air at temperatures ranging from 1250 °C to 1500 °C, for 1 h. The densities of the sintered samples were measured using either weight and dimensions or Archimedes method. The chemical analysis was determined using a XRF (JEOL, JSM3200). For the conductivity measurements the sintered specimens were painted with platinum paste and heat treated at 900 °C for 1 h to make two electrodes. The AC conductivity measurements were conducted using an impedance analyzer (4192 A, LF Hewlett & Packard — 5 Hz to 13 MHz) in air at temperatures from 400 °C to 500 °C. The microstructure investigation was performed using a scanning electron microscope SEM (JEOL, JSM-5400, Japan).

3. Results and discussion

3.1. Densification of (8YSZ+xCuO) compacts

Table 1 shows the sintered densities in percentages of the theoretical density of the 8YSZ, neglecting the effect of the CuO, since it was found by XRF analysis – before and after sintering – that the amount left of CuO in the sintered samples as $Y_2Cu_2O_5$ was almost one quarter of its initial value [7]. It can be seen from the table that the samples containing CuO have higher sintered densities relative to the samples of 8YSZ. The samples containing 0.3 mol% CuO showed eventually the highest sintered densities.

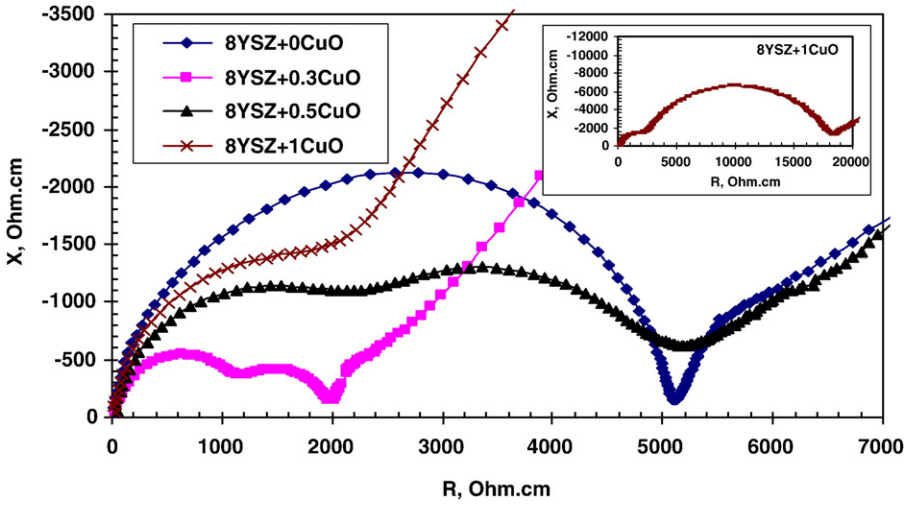

Fig. 2. Impedance spectroscopy for 8YSZ+xCuO sintered at 1250 °C/1 h and tested at 500 °C.