Resistance Oscillations in Doped $BaCeO_3$ and $BaZrO_3$ in Water Containing Atmospheres

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INTRODUCTION

Barium cerate ($BaCeO_3$), when doped with a lower valent ion on the Ce-sublattice, becomes oxygen-deficient with oxygen vacancies as the predominant point defects, and exhibits oxygen ion as well as electronic conduction (1-3). When water is dissolved into the structure to fill up oxygen vacancies, it is known to exhibit protonic conductivity (3,4). Many perovskites are known to exhibit similar behavior. Conduction properties are usually studied using AC impedance technique. In this work, transport properties of doped $BaCeO_3$ and $BaZrO_3$ were investigated using a four-probe DC technique in a constant current mode. It was observed that the resistance of the sample exhibits periodic oscillations in water-containing atmospheres. To the authors' knowledge, such oscillations have not been reported in this material, or other perovskites. The objective of this paper is to present the experimental observations on oscillations, and present plausible reasons for the observed effects.

EXPERIMENTAL

Dense ceramic samples of Gd-doped barium cerate and Y-doped barium zirconate were made by conventional solid state method, comprising calcinations, powder compaction, and sintering in air (5). The sintered specimens were then ground into rectangular bars (about 4 cm x 1cm x 0.2 cm). Platinum or gold paste was printed on the sample surface, followed by heating at 850°C for three hours to render strong bonding with the ceramic. Platinum or gold wires were used as the leads for electrical measurement. Resistance measurements were performed using a 4-probe DC method at several temperatures and in dry and wet atmospheres. Water vapor was supplied by bubbling water with a dry carrier gas, and water partial pressure was varied, by varying the bubbler temperature. In each isothermal experiment, resistance was recorded as function of time in dry and wet atmospheres .

RESULTS AND DISCUSSION

Figure 1 shows one of the several resistance vs. time traces, measured with platinum electrodes. The data were recorded at 900°C with a bubbler temperature of 40°C. It can be seen that oscillations occurred only in a water-containing atmosphere. The period of oscillations was typically several tens of minutes to a couple of hours. By contrast, in dry atmospheres, both air and nitrogen, no oscillations were observed. This suggests that oscillations are induced by water vapor. Also, no oscillations were observed for a bubbler temperature of 25°C, indicating that oscillations do not occur at too low a vapor pressure of water. Similar behavior was also observed with gold electrodes, indicating that the phenomenon is not specific to the electrodes used. Figure 2 shows resistance vs. time traces with gold electrodes. In all experiments, the temperature was maintained constant, to within a couple of degrees. This excludes the possible role of temperature fluctuations as the source of oscillations, as determined on the basis of known activation energies for transport. Possible reasons for the observed oscillations will be discussed.

ACKNOWLEDGEMENTS

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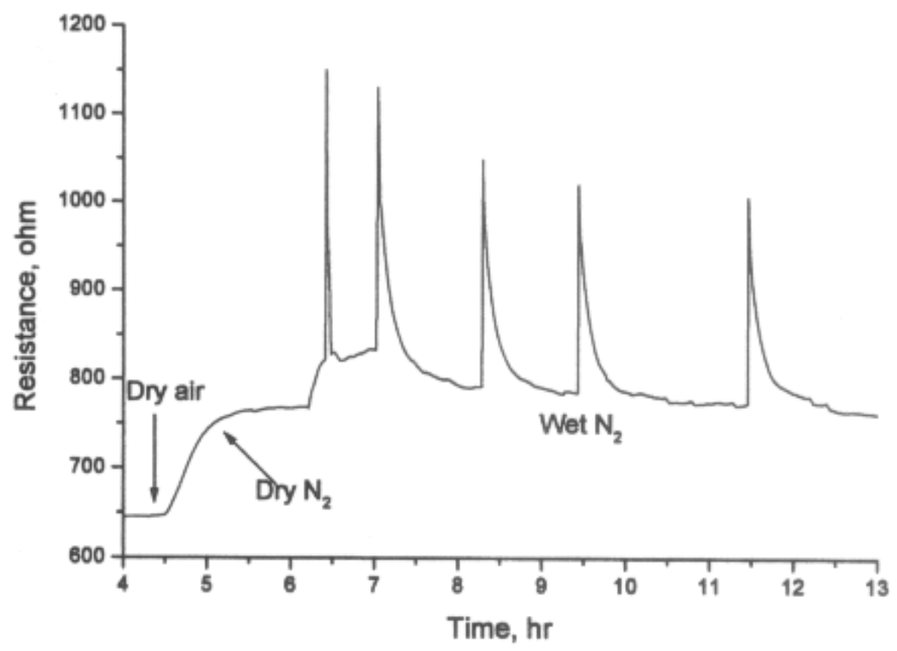

Figure 1. Resistance vs. time at 900°C with a bubbler temperature of 40°C: Platinum electrodes.

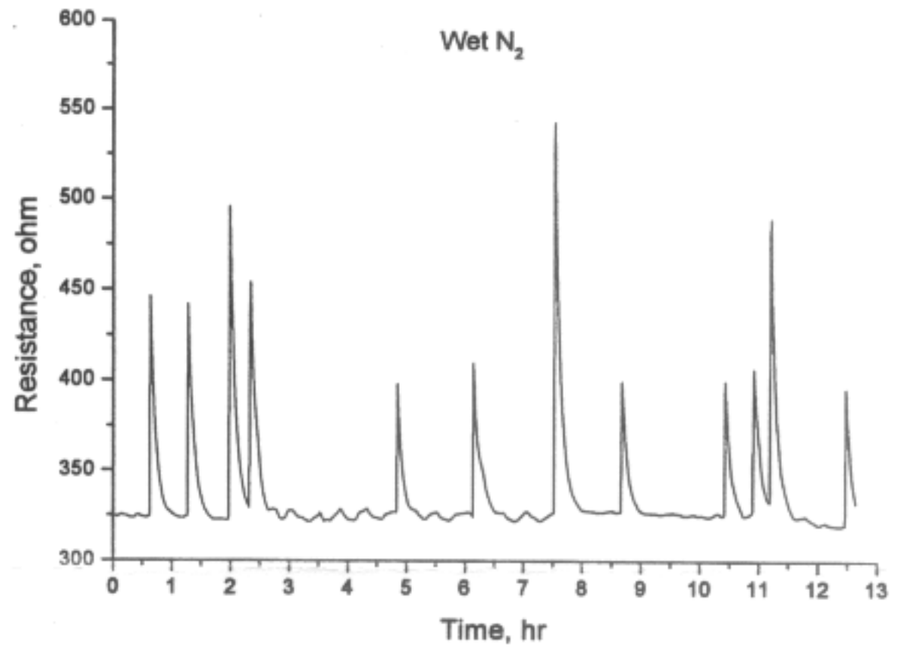

Figure 2. Resistance vs. time at 900°C with a bubbler temperature of 55°C: Gold electrodes

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