Chemical Stability and Electrical Conductivity of Proton Conducting Perovskites under High H_2O Pressures

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INTRODUCTION

Proton conducting perovskites have potential applications to intermediate temperature solid oxide fuel cells, hydrogen separation, and hydrogen production by the decomposition of H_2O [1]. Among them, Y-doped $BaZrO_3$ and $Ba_3Ca_{1.18}Nb_{1.82}O_{9-\delta}$ (BCN18) have been shown to exhibit superior proton conductivity and stability against CO_2 and water vapor. Nevertheless, their proton conductivity is not maximized due to the low H_2O pressures examined. When exposed to a high H_2O pressure, proton concentration should be increased, and the mobility of protons may be enhanced due to extremely low concentration of oxygen vacancies. To produce hydrogen from H_2O decomposition, materials have to be exposed to a pressurized steam, under which conditions the chemical stability of the materials is not known. Recently, electrical conductivity and chemical stability of $BaZrO_3$ and BCN18 in the presence of pressurized steam were investigated at various temperatures. The experimental procedure used and results obtained will be described.

EXPERIMENTAL

Dense and porous materials of composition $Ba_3Ca_{1.18}Nb_{1.82}O_{9-\delta}$ (BCN18) and $BaZr_{0.93}Y_{0.07}O_{3-\delta}$ (BZY7) were prepared using conventional procedures as described in [2, 3]. Platinum paste was painted on dense and porous samples to form four contacts in the form of strips. Four platinum wires were attached to each sample to form four leads for dc four-probe measurements. Samples were placed in a tightly sealed autoclave, where H_2O pressure (up to ~325 bars) was generated by adding a prescribed amount of H_2O and heating to a high temperature. The temperature, H_2O pressure, and electrical conductivity were monitored by a computerized data acquisition system. The samples after exposure to steam were characterized using X-ray diffraction to examine if decomposition of the materials occurred.

RESULTS AND DISCUSSION

Samples of BZY7 and BCN18 were exposed to pressurized steam at p_{H_2O} from 9 to 325 bars at a constant temperature of 500^{o}C for ~ 10 hours. Figure 1 shows the XRD of BZY7 after exposure to p_{H_2O} =100 bars. It is seen that new peaks (labeled "N") are present in addition to the peaks due to BZY7. The peaks at 2θ=24.89 and 28.79^o correspond to those of barium hydroxide, which is likely to be one of the decomposition products. Further efforts are underway to identify the decomposition products. Similar results were obtained on BCN18, as shown in Figure 2. It appears that both materials are unstable at p_{H_2O} higher than nine bars. It was also observed that dense BCN18 broke during the experiment while porous BCN18 and BZY7 did not. Further details on this subject and electrical conductivity measurements will be reported.

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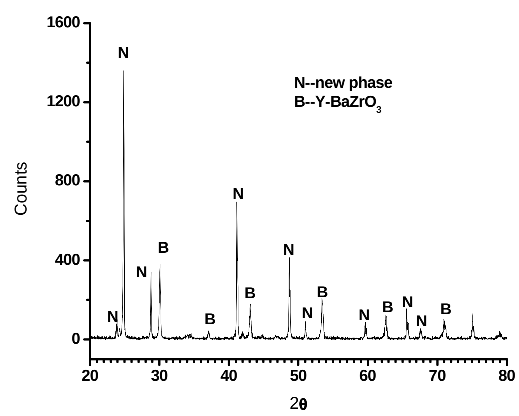

Figure 1. X-ray diffraction trace of $BaZr_{0.93}Y_{0.07}O_{3-\delta}$ after exposure to steam at p_{H_2O} =100 bars and at 500^{o}C

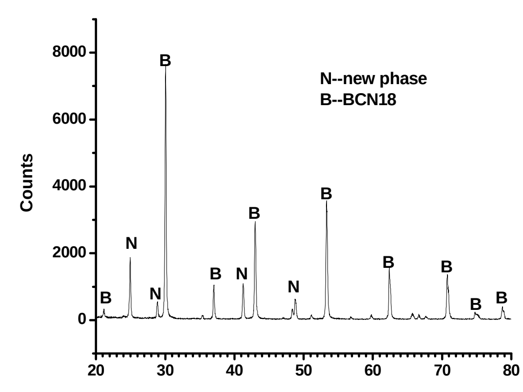

Figure 2. X-ray diffraction trace of BCN18 after exposure to steam at p_{H_2O} =40 bars and at 500^o