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Enhanced sulfur tolerance of $BaCe_{0.65}Zr_{0.20}Y_{0.15}O_{3-\delta}$-$Ce_{0.85}Gd_{0.15}O_{2-\delta}$ composite for hydrogen separation membranes

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Abstract

Thanks to its high hydrogen permeability and good chemical stability in moist CO_2 environments, $BaCe_{0.65}Zr_{0.20}Y_{0.15}O_{3-\delta}$-$Ce_{0.85}Gd_{0.15}O_{2-\delta}$ mixed conducting material is considered one of the most promising candidates for hydrogen separation ceramic membranes. In this work, its chemical stability under H_2S-rich atmosphere was systematically investigated by in-situ electrochemical characterizations and ex-situ structural, chemical and morphological analyses. A performance degradation of the total conductivity depending on the H_2S content was observed: at 700 °C and under 1500 and 700 ppm of H_2S the conductivity drop was 15% and 2% respectively. The complementary information gathered by morphological and chemical analyses showed that the changes responsible for the total conductivity degradation are mainly confined to the surface of the membrane. Indeed, after the exposure to the H_2S-containing atmosphere, some traces of sulfur-related compounds were detected only on the top of the membrane while the bulk preserved a fully dense structure with well-defined grain boundaries and no evidence of cracks. However, no evidence of S-based compounds were revealed by structural investigations, probably due to the detection limit of these techniques and/or to the low crystallinity of the secondary phases. Contrary to Pd-based membranes that are severely deteriorated by a few ppm of sulfur, this material shows an acceptable stability even under 700 ppm of H_2S and could be attractive for tailored applications such as, for example, operations related to steam reforming of methane often containing 10-300 ppm of H_2S.

Keywords: composite membrane, hydrogen separation, chemical stability, H_2S-rich atmosphere

Introduction

Materials based on mixed proton-electron conductors (MPEC) are currently attracting growing interest for their potential applications in H_2 gas separation membranes or in catalytic membrane reactors (CMR).[1-5]