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Anodic polarization induced performance loss in $GdBaCo_2O_{5+\delta}$ oxygen electrode under solid oxide electrolysis cell conditions

Bo Wei^{a,*}, Jiebing Feng^a, Lin Zhu^a, Zhihong Wang^a, Xingbao Zhu^a, Xiqiang Huang^a, Yaohui Zhang^a, Lingling Xu^{b,*}, Hong Gao^b, Zhe Lü^a

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ABSTRACT

Double perovskite oxide $GdBaCo_2O_{5+\delta}$ (GBCO) is widely investigated as promising cathode for solid oxide fuel cells (SOFCs), but it remains unclear whether GBCO is suitable for application in solid oxide electrolysis cells (SOECs) anode. In this study, the effect of anodic polarization on electrochemical activity and microstructure of GBCO electrode are investigated under SOECs operation conditions. Both polarization and impedance spectra results clearly demonstrate that anodic bias treatment leads to substantial performance degradation and higher anodic current passage causes more serious activity loss. The deactivation behavior can be mainly ascribed to the formation of surface BaO precipitates in harsh oxidation atmosphere, as revealed by microstructural observation. Our study suggests that GBCO oxide is not suitable for SOECs anode application, but significant change of surface chemistry enables it a good model electrode for segregation related studies.

1. Introduction

Solid oxide electrolysis cells (SOECs) offer great potential for efficient and economic production of fuels (e.g. hydrogen or syngas) from water electrolysis or co-electrolysis processes [1–4]. SOECs can thus sesrve as an alternative tool for energy storage from various renewable sources, particularly for intermittent wind or solar energy [5]. Compared to conventional low temperature water electrolysis, high operation temperature of SOEC is thermodynamically advantageous, because both the required electrical power at higher temperature and the electrode overpotentials are reduced [3,5].

In principle, SOECs are reversely operated solid oxide fuel cells (SOFCs). Therefore, great achievement on materials and technologies in SOFCs can be directly applicable to the development of SOECs. As for oxygen electrode (anode), (La,Sr)MnO₃ (LSM) is the most commonly used anode for the oxygen evolution reactions (OER) of SOECs, but the performance is largely limited by its poor ionic conductivity [2,3]. Alternative oxygen electrodes with mixed ionic and electronic conductivities are continuously being developed and excellent candidates like $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3.8}$ (LSCF), $La_{0.6}Sr_{0.4}CoO_{3.8}$ (LSC) and $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3.8}$ (BSCF) electrodes are widely studied with higher performance [6–10].

Similar to SOFCs, the performance and efficiency of SOECs are

critically dependent on the electrocatalytic activity and stability of oxygen electrodes. However, long term degradation of oxygen electrode in electrolysis mode often occurs faster than in fuel cell mode [5]. At the oxygen electrode side, the physical, chemical and microstructural changes of the electrode and interface regions will occur under SOECs operation conditions which can contribute to the degradation of the cell/stack. For example, the delamination of LSM electrode from YSZ electrolyte and disintegration of electrode particles at the interface region have been observed in LSM based electrodes [11–13]. While for LSCF based electrodes, phase change and pronounced SrZrO₃ formation have been observed [9,14,15]. Moreover, the poisoning of extrinsic contaminants like gaseous chromium species from Fe-Cr metallic alloys can also lead to performance loss on oxygen electrodes [16–18].

Recently, double perovskite oxides of REBaCo₂O_{5+ δ} (RE = Pr, Nd, Gd, Sm etc.) have won tremendous attention as promising cathode materials for reduced temperature SOFCs [19–26]. The ordered Ba cations in the A-site and the localized oxygen vacancies in the rare-earth plane (REO_x) can result in very fast oxygen-ion transport (D^*) and surface exchange coefficients (k^*), which are very advantageous for oxygen reduction reactions. Tarancon et al. and Kim et al. have reported, respectively, promising D^* and k^* values for GdBaCo₂O_{5+ δ} (GBCO) and PrBaCo₂O_{5+ δ} (PBCO) compared with other types of perovskite or perovskite-related oxides [21,23]. For example, the D^* and

E-mail addresses: bowei@hit.edu.cn (B. Wei), xulingling hit@126.com (L. Xu).

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^a Department of Physics, Harbin Institute of Technology, Harbin, 150080, China

b Key Laboratory of Photonic and Electric Bandgap Materials, Ministry of Education, School of Physics and Electronic Engineering, Harbin Normal University, Harbin, 150025, China

^{*} Corresponding authors.

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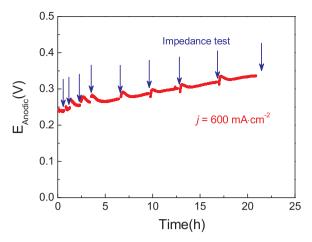
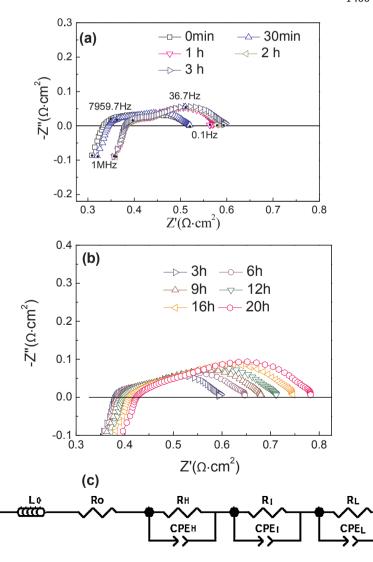


Fig. 1. Anodic polarization curve (η -t) of GBCO electrode as a function of time. The electrode was measured under anodic current passage of 600 mA cm $^{-2}$ at 750 °C in air.

 k^* values at 500 °C are about 3×10^{-10} cm 2 s $^{-1}$ and 8×10^{-8} cm s $^{-1}$, respectively, much higher than 1.2×10^{-12} cm 2 s $^{-1}$ and 2×10^{-8} cm s $^{-1}$ for state-of-the-art LSCF at the same temperature [21]. These encouraging results have motivated many research groups to focus on the development of high performance cathodes based on these double-perovskite oxides. Excellent electrochemical activity



towards oxygen reduction reactions at intermediate temperatures have been reported [19–24]. Unfortunately, these cobalt-based cathodes also suffer from a serious problem of high thermal expansion coefficients (TECs, $\sim\!20\times10^{-6}\,\mathrm{K^{-1}}$) [19,21], which are about twice larger than that of YSZ and ceria based electrolytes (10–13 \times 10 $^{-6}\,\mathrm{K^{-1}}$). This large mismatch could be mitigated through the design of functionally graded architecture [26]. Furthermore, Druce et al. have found that Ba element from GBCO lattice segregates strongly to the outermost surface with the formation of a Ba-rich layer, which probably could impact the activity and stability of oxygen electrode [20].

To date, although the performance of REBaCo $_2$ O $_{5+\delta}$ oxides as SOFC cathodes has been widely investigated, there is very limited study about SOEC anode using these layered electrodes [25]. In the present study, the electrochemical performance and microstructural change of GBCO anode were investigated under different current passages at 750 °C in air. The possible degradation mechanism of the electrode activity in SOEC condition was discussed.

2. Experimental

2.1. Fabrication of half-cells

To prepare dense electrolyte pellets, sol-gel process derived $\rm Sm_{0.2}Ce_{0.8}O_{1.9}$ (SDC) powder ($\sim\!0.25\,\rm g$ for each) was uniaxially pressed into pellets with a diameter of 13 mm and followed by sintering at 1400 °C for 4 h. The thickness of as-obtained SDC substrates was

Fig. 2. (a, b) Impedance responses for the oxygen evolution reaction on the GBCO oxygen electrode. These spectra were collected under open circuit condition during the anodic current passage of $600\,\mathrm{mA\,cm^{-2}}$ at $750\,^\circ\mathrm{C}$ in air, (c) an equivalent circuit for the fitting of the impedance data.

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