INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2017) 1–7

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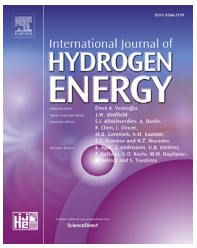

Quantitative assessment of anode contribution to cell degradation under various polarization conditions using industrial size planar solid oxide fuel cells

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ARTICLE INFO

Article history:
Received 27 August 2017
Received in revised form
6 November 2017
Accepted 4 December 2017
Available online xxx

Keywords:
Ni-YSZ anode
Polarization
Degradation
Quantitative assessment
Solid oxide fuel cell

ABSTRACT

Quantitative assessment of anode contribution to cell performance has been investigated under various polarization in three stack repeating units made of 10 cm × 10 cm planar anode-supported solid oxide fuel cells. The measurement is performed in-situ by inserting an ultra-thin Pt probe at the anode/electrolyte interface. The results reveal that the polarization resistance accounts for more than 90% of the total cell resistance when the cell is operated under the activation and concentration polarizations, respectively. The anode almost has no effect on cell degradation when the cell is operated under activation polarization. However, the anode has an obvious contribution to the cell degradation when the cell is operated under ohmic and concentration polarization, where the anode voltage increases by 23.36%/100 h and 512.28%/100 h, respectively. The anode operated under concentration polarization has twenty times of contribution to cell degradation than that of the ohmic polarization. However, when the cell is operated under the ohmic polarization, the main degradation comes from the ohmic resistance caused by the anode.

Introduction

Solid oxide fuel cell (SOFC) is an efficient energy converting device which can generate electricity directly from electrochemical reaction [1]. The SOFCs have great potentials for energy applications, such as combined heat and power (CHP) generation, auxiliary power unit (APU), range extender for electric vehicles [2–4] due to the advantages of high efficiency, fuel adaptability, and so on [5,6]. Typical SOFC consists of anode, electrolyte, cathode, interconnect and sealing [7,8]. The anode provides reaction sites for the electrochemical reaction and pathways for electrons, in which Ni-YSZ cermet is considered as one of the most mature anode materials [9–11].

In a typical performance curve of SOFCs [12], the curve is generally composed of three polarization zone, including activation, ohmic and concentration polarization [12,13]. The polarization loss is closely related to the current density. The

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https://doi.org/10.1016/j.ijhydene.2017.12.030

activation polarization happens in low current, ohmic loss is inversely proportional to the current. The resistance keeps constant during ohmic polarization. The concentration polarization exists during the whole discharging process, and obviously under high current due to lacking adequate gaseous reactants into the reaction sites. The loss of ohmic polarization, activation polarization and concentration polarization has strong dependence on the cell structure and significantly determines the cell output performance [14–17]. As a key component of the single cell, the electrochemical properties and mechanism of polarization process of the anode are mainly dependent on its composition, particle size and porosity, etc [16–23].

The anode electrochemical reaction mainly takes place at the triple-phase boundary region of the anode/electrolyte interface and determines the cell performance. Much work has been conducted to investigate the effect of the anode on cell performance [24–26], but most of them did not get the anode contribution quantitatively, because it is very difficult to insert voltage probe on the anode/electrolyte interface. Thus, it is unclear on the anode contribution to the overall cell performance during cell operation and it makes a challenging on how to improve the anode. In this work, an ultra-thin voltage probe was inserted in the anode/electrolyte interface based on the method developed in our previous work [27,28], and electrochemical properties of the anode and its contribution to cell performance were quantitatively investigated in-situ. In addition, it can be seen that most of work related to effect of anode on cell performance was conducted only by button cell and focused on that the cell run in ohmic polarization [29,30]. While in fact, the cell will meet complex condition, such as shortage of fuel, or suffering fuel starvation, etc, in industrial application. This work aims to investigate the anode on cell performance under various polarization by industrial-sized cell through our designed method mentioned above. It will be very helpful to improve the application of cell and provide reference to optimize its anode structure for planar solid oxide fuel cells.

Experimental

To investigate the effect of anode on cell performance in situ in a large-scale stack, an industrial sized cell with Pt probe in anode/electrolyte interface was designed and prepared. In this work, Ni-YSZ/YSZ/GDC/LSCF-GDC cells were chosen. The cell composition, such as LSCF, GDC, etc., can be found in our previously published work [27]. To prepare the designed cells, the anode tape was firstly prepared by tape casting from a NiO-YSZ slurry with a thickness of about 120 μm. The slurry consisted of 45 wt% NiO and 55 wt% YSZ. Four layers of anode tapes and a 10 μm-thick active anode tape were then laminated together to form the anode, during which a Pt strip was printed onto the surface of the tape with size of 2 cm × 0.5 cm as an internal voltage probe with a thickness ~2 μm, where its position is close to the middle of the cell between fuel in and fuel outlet. YSZ slurry was subsequently sprayed onto the anode followed by sintering at 1320 °C for 4 h. A 5 μm-thick barrier layer of GDC was then sprayed onto YSZ and sintered at 1320 °C for 1 h. Finally, a 30 μm thick active cathode was sprayed and sintered at 1050 °C. To measure the cell performance, Pt probes were printed on the surface of both anode and cathode. Furthermore, in order to ensure an intimate interface between the electrodes and the interconnector, a 30 μm thick NiO and a 130 μm thick LSCF were screen-printed on the anode and cathode side of the full cell, respectively. The procedure mentioned above is shown in Fig. 1. In Fig. 1 b, the value between probes 3 and 2 during DC discharging means the voltage produced by the anode, and the value between probes 2 and 1 means the voltage produced by the electrolyte and the cathode.

After the cells were prepared, three stack repeating units (SRUs, one cell and two interconnectors) were assembled separately using three anode-supported industrial-sized cells with 100 cm^2 total cell area and 63 cm^2 active area. The detailed assembling process of the SRU can be found in our previous work [31–33]. The three cells in the three SRUs were numbered 1, 2 and 3, respectively, i.e. SRU 1 and cell 1, SRU 2 and cell 2, SRU 3 and cell 3. The SRU was heated to 900 °C with a rate of 1 °C min^{-1}, held for 4 h, and then cooled to 750 °C. Prior to the electrochemical measurement, pure N_2 was fed into the anode for 10 min. Then, 0.5 SLM dry hydrogen was fed into anode to reduce the anode for 30 min. The cathode side was fed with 1.5 SLM air as the oxidant gas. After the anode was completely reduced, the electrochemical performances of cell and anode were obtained by DC and AC measurements. The three individual cells in the three SRUs were operated for 200 h under activation polarization, concentration polarization and ohmic polarization conditions, respectively. SEM (FEI Quanta FEG 250 with EDS) was employed to characterize the cell microstructure.

Results and discussions

The I-V curves of the cells 1, 2, and 3 in the corresponding SRUs were shown in Fig. 2 a. The performance of cell 2 is close to that of cell 1, and both of them are slightly higher than that of cell 3. To distinguish the polarization zone, the I-V curves ware treated by a first differential ($\frac{dV}{dI}$, means resistance) and a second differential ($\frac{dR}{dI}$, means resistance changes with current), as shown in Fig. 2 b. It demonstrated that the operating curves changed dramatically from 0 to 10 A, and then stayed stable during 10–50 A, and obviously fluctuated again when the current increased to more than 50 A. Accordingly, the operating curve can be separated to three stages, where the first stage is the active zone below the current of 10 A, the second stage is the ohmic zone under the current between 10 and 50 A and the last is the concentration zone with the current above 50 A. Therefore, to make it clearer, the constant current of 3 A, 62 A and 30 A was chosen as the discharging current for long-term operation as activation, concentration and ohmic polarization, respectively, for the cell 1, 2 and 3 in the three SRUs.

Fig. 3 showed the performance decreasing with operating time of the cell and the anode, respectively of the individual cells in three SRUs. It can be determined that the voltages of cells 1, 2, and 3 decreased with degradation rate of 0.44%/100 h, 4.38%/100 h, and 4.36%/100 h, respectively. The output performance degradation rate of cell 1 operated under the

Please cite this article in press as: Yu R, et al., Quantitative assessment of anode contribution to cell degradation under various polarization conditions using industrial size planar solid oxide fuel cells, International Journal of Hydrogen Energy (2017), https://doi.org/10.1016/j.ijhydene.2017.12.030