Highly Redox-Resistant Solid Oxide Fuel Cell Anode Materials Based on La-doped SrTiO_3 by Catalyst Impregnation Strategy

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An anode backbone using 40wt% $(\text{ZrO}_2)_{0.89}(\text{Sc}_2\text{O}_3)_{0.1}(\text{CeO}_2)_{0.01}$ (SSZ)- $\text{Sr}_{0.9}\text{La}_{0.1}\text{TiO}_3$ (SLT) cermet was prepared for SSZ electrolyte-supported SOFC single cells. 15 mgcm^{-2} $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_2$ (GDC) was impregnated to totally cover the SSZ-SLT anode

backbone surface acting as a catalyst, and the cell voltage achieved 0.865V at 200 mAcm⁻² using (La_{0.75}Sr_{0.25})_{0.98}MnO₃ (LSM)-SSZ cathode in 3%-humidified hydrogen fuel at 800°C. Cell performance was substantially improved from 0.865V to > 0.97V when 0.03 mgcm⁻² Pd or Ni was further incorporated as a secondary catalyst into the anode layer. 50 redox cycles were performed to investigate redox stability of this high performance anode. It was found that even after the 50 redox cycle long-term degradation test, cell voltage at 200 mAcm⁻² was retained around 0.94V, higher than the cell performance using the conventional Ni-SSZ cermet anode. The catalytically-active reaction sites at ceria-Pd or ceria-Ni may account for the excellent performance, and the extremely low metal catalyst concentration prevent serious metal aggregation in achieving excellent redox stability.

Keywords:

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1. Introduction

The porous Ni-Zirconia cermet has been developed for decades as the state of the art SOFC anode because of its high performance and low cost. To achieve sufficient current collection and catalytic activity, the nickel content is usually over 35 vol.% to form a connecting path for electron transport [1].

However, nickel in the anode could be oxidized in the downstream part of fuel flow in which a large amount of steam is contained at a high fuel utilization. The oxidation and reduction of Ni in the vicinity of triple-phase boundary (TPB) will cause anode operation instability or even irreversible deterioration [2]. Moreover, the cyclic reduction and oxidation (redox cycling) of the anode is likely to occur in a commercial SOFC system due to situations such as inhomogeneous fuel flow, seal leakage, fuel supply failure, and/or system shutdown. The reduction and oxidation of Ni will cause large volume changes, which would lead to fracture either of the anode itself or of its adjacent component, the thin electrolyte layer [3-6].

Oxide anode has been drew much attention for years. Perovskite materials such as doped LaCrO_3 [7, 8], doped SrTiO_3 [9], and the double perovskite materials $\text{Sr}_2\text{Mg}_{1-x}\text{Mo}_x\text{O}_{6-\delta}$ [10] have been developed as SOFC anodes. However, the electronic

conductivity of doped LaCrO_3 is only on the orders of 1 Scm^{-1} under SOFC anode conditions [11], and the double perovskites are reported not stable in reducing atmosphere above 900°C [12].

Marina et al. [9] and Wang et al. [13, 14] thoroughly evaluated the electrical and thermal-mechanical properties of $\text{Sr}_{1-x}\text{La}_x\text{TiO}_3$ as a possible SOFC interconnect material. According to Wang's reports, the air-sintered 10% La-doped SrTiO_3 has a thermal expansion coefficient (TEC) of $11 \times 10^{-6} / ^\circ\text{C}$ and an acceptable electrical conductivity ($\sim 13 \text{ S/cm}$) under the SOFC anode condition. In addition, the air-sintered $\text{Sr}_{0.9}\text{La}_{0.1}\text{TiO}_3$ (SLT) shows negligible linear expansion in the cyclic redox test.

Impregnation method is a widely accepted approach to improve SOFC electrochemical performance because of the well-dispersed nano-sized catalysts formed by thermal decomposition, and to improve redox stability due to structural advantages [15-18]. Recently, some groups focused the stability of the catalyst impregnated SOFC anode. Miller *et al.* investigated the $\sim 550\text{h}$ test of Ni impregnated anode at open circuit voltage condition, and found the Ni nano-particles coarsening after measurement [19]; Ramos *et al.* compared stability of the Ru/GDC and Ni/GDC impregnated anode, and found Ru/GDC impregnated anode has better degradation resistance in highly humidified hydrogen [20]; Zhang *et al.* confirmed the instability and growth of

impregnated Ni/GDC under a variety of H₂O/H₂ conditions by TEM, and concluded the morphological change negatively affected the electrochemical performance [21]; Fan *et al.* observed the redox stability of the GDC impregnated anode during 11 redox cycles, however, their cell performance was not comparable with conventional Ni-SSZ anode [22]. To our limited knowledge, only a few papers systematically investigated the redox stability of the catalyst impregnated SOFC anode, and no publication proposed a satisfactory strategy for SOFC anode to achieve both high performance and excellent stability against the detrimental frequent redox cycling.

In this study, we utilize the stable 40wt% SSZ - SLT composite to fabricate a robust SOFC anode backbone. Then the first catalyst phase 15 mgcm⁻² Ce_{0.9}Gd_{0.1}O₂ (GDC) will be incorporated into anode by a wet impregnation method, and then 0.03 mgcm⁻² metal catalyst such as Pd or Ni will be impregnated into the anode as a secondary catalyst phase. Cell performance and long-term redox cycling stability are investigated.

2. Experimental

2.1. Cell Fabrication

Scandia-stabilized zirconia (SSZ) plates (10mol% Sc_2O_3 - 1 mol% CeO_2 - 89 mol% ZrO_2 , Daiichi Kigenso Kagaku Kogyo Co., Ltd., Osaka, Japan) with a diameter of 20 mm and a thickness of 200 μm were used as solid electrolytes. $\text{Sr}_{0.9}\text{La}_{0.1}\text{TiO}_3$ (Praxair, Inc., Connecticut, US) powder was mixed with 40wt% SSZ, and then the composite powder was further mixed with 20wt% carbon black (Sigma-Aldrich Co.) as pore former. The well mixed powder was homogeneously blended with organic binder (α -terpineol) as the anode functional layer pastes. 50wt% LSM (Praxair, Inc., Connecticut, US) powder was blended with SSZ in the binder as the cathode functional layer paste. The anode current collecting paste and the cathode current collecting paste were prepared by mixing the binder with $\text{Sr}_{0.9}\text{La}_{0.1}\text{TiO}_3$ (with 20wt% carbon black as pore former) and LSM, respectively. The anode functional layer (30 μm in thickness, 0.64 cm^2 in area) was screen printed on the electrolyte plate. After drying, the second and the third ~ 30 μm $\text{Sr}_{0.9}\text{La}_{0.1}\text{TiO}_3$ current collecting layers were screen printed, followed by sintering in air at 1300°C for 3 h. The LSM/SSZ (1-to-1 mixture in weight) composite functional layer, the second and the third LSM conducting layers were screen printed on the counter side of the SSZ electrolyte plate, followed by sintering in air at 1200°C for 5 h. As for the impregnation, the well-sintered single cells were impregnated with 4 μl aqueous solution of $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ and $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (1-to-9 mixture in

mole) on the top of the anode, then the solution was automatically filled into porous anode by capillary force. We then dry and heat the impregnated cell at 800°C for 1h in air in order to form Gd-doped CeO₂. The impregnation process was repeated 8 times to achieve the GDC loading of 15 mgcm⁻². It should be noted that the repetition process depends on the concentration of metal nitrate solution and target catalyst loading, so that repetition could be variable depending on each situation. In addition, dilute Ni(NO₃)₂·6H₂O or Pd(NO₃)₂ (Kishida Chemical Co., Ltd.) solution was also impregnated into the anode as the secondary phase catalyst to achieve Ni or Pd loading of 0.03 mgcm⁻² using the same impregnation process, followed by thermal decomposition at 800°C for 1h. 1g 40wt% SSZ-SLT oxide powder was mixed with 0.25g carbon black, and the mixed powder was pressed and sintered at 1300°C for 3 h as a porous SSZ-SLT pellet. The porous SSZ-SLT pellet was impregnated as high as 1g GDC using the same process for the impregnation of anode; after GDC impregnation and thermal-treatment at 800°C for 1h, the pellet was grinded into powder for X-ray diffraction measurement (XRD, Rigaku Smart Lab) to identify the formation of the Gd-doped CeO₂. The microstructure of the anode was investigated using FESEM-EDX (Hitachi S-5200).

Pt mesh was attached to each electrode surface as a current collector using SLT and LSM slurry as paste avoiding any direct contact between Pt mesh and electrode functional layers. Pt paste was used as a reference electrode (RE) locating more than 2 mm away from the edge of the cathode.

2.2. Cell Test

To measure the electrochemical performance, the single button cell was sandwiched by two alumina rings and sealed by ceramic binder, and then the cell was attached to alumina tubes and sealed with a glass sealant [23-25]. The cell was tested at 800°C with 100 mLmin⁻¹ 3% -humidified H₂ as the fuel and 100 mLmin⁻¹ air as the oxidant. Prior to the cell testing, the anode was exposed in the hydrogen atmosphere at 900°C for 2 h to reduce NiO or PdO. The cell performance was evaluated by the Fuel Cell Test Station (Toyo Corp.). The voltage loss on the anode side was measured from the potential difference between the anode and the reference electrode (on the cathode side) and the open circuit voltage. The anode-side voltage loss was then divided into the anodic overvoltage and the anode-side ohmic loss using the well-known current interruption technique. These non-ohmic and ohmic contributions were experimentally separated by

regularly monitoring the voltage relaxation at 15 μ s, just after current interruption [23]. Within 15 μ s, ohmic voltage loss due to ionic and electronic conduction can effectively be relaxed [24]. Most of the (non-ohmic) electrode processes have longer relaxation time, i.e., lower relaxation frequencies [24]. Note that the non-ohmic anodic overvoltage may consist of activation polarization and concentration polarization, the separation of which may be possible in a future study, by applying e.g. impedance analysis.

2.3. Redox cycling test

The redox cycling test was performed based on the SOFC shutdown protocol for the real SOFC systems published previously [25]. In this protocol, the fuel supply shuts off and the temperature changes very quickly, the cell was cooled down to 400°C in 1h, and then maintained at 400°C for 30 min. The cell was then heated to 800°C in 1h, and kept for 1h. Fuel was supplied during heating and power generation; however, the fuel supply was stopped during cooling and at the lowest temperature. After each cycle, the temperature was maintained at 800°C, and the cell voltage was measured at a constant current density (0.2 Acm⁻²) [25]. In this study, we only utilized the fuel interruption based on the shutdown protocol to perform redox cycles and kept the cell at the constant

temperature of 800 °C, while other parameters maintained the same as the shutdown protocol.

3. Results and discussion

3.1. Pure oxide anode

The XRD patterns of the GDC-impregnated 40wt%SSZ-SLT composite powder was shown in Figure 1. The Bragg peaks of GDC were identified using MDI jade 6.5 (XRD analysis software

Figure 1. The XRD patterns of $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_2$ -impregnated SSZ-SLT composite.

installed). No Bragg peaks of Gd_2O_3 were detected indicating that the GDC was synthesized in the thermal decomposition process within the porous 40wt%SSZ-SLT structure. The synthesis of GDC in the porous anode would be similar to our test in the porous pellet, where more GDC could be included in the porous pellet than the porous anode. We therefore concluded that the GDC could be achieved during the anode impregnation process.

Performance of the 40wt%SSZ-SLT composite anode and the GDC-impregnated 40wt%SSZ-SLT composite anode was investigated by I-V measurement shown in Figure 2. GDC as an effective catalyst significantly promoted cell performance by substantially reducing the anodic overvoltage. The anodic overvoltage of the

Figure 2. Anode-side ohmic loss, anodic overvoltage, I-V curve and I-P curve of SOFC single cell using (a) the 40wt% SSZ-SLT anode and (b) the GDC-coated 40wt% SSZ-SLT anode.

40wt%SSZ-SLT composite was reduced from 0.41 V to 0.04V at 50 mAcm⁻² after GDC was incorporated. While the impregnated GDC may partially act as a mixed ionic and electronic conductor around the anode/electrolyte interface, here we considered that the improvement of cell performance came mainly from a decrease in anodic overvoltage due to the catalytic effect of GDC because of the well-known redox properties [26]. However, the cell performance was comparatively lower than that using the conventional Ni-SSZ anode that contains around 50 wt.% Ni inside, acting as both superior catalyst for hydrogen oxidation and electronic conducting path [25].

In this study, the impregnation of 15 mgcm⁻² GDC reached the maximum load of our porous 40wt%SSZ-SLT anode backbone as indicated in Figure 3 (a), the anode side

Figure 3. Cross-section on (a) the anode side and (b) the cathode side of the SOFC single cell using GDC-impregnated 40wt% SSZ-SLT anode.

cross-sectional SEM micrograph of the SOFC single cell. Compared with the cathode side as shown in Figure 3 (b), the porosity of the anode was lower than that of the cathode due to the large amount of the GDC impregnation, and further impregnation into the anode was not easily accessible. The microstructure of the porous 40wt%SSZ-SLT anode backbone and the GDC-impregnated 40wt%SSZ-SLT anode was shown in Figure 4 (a) and Figure 4 (b), respectively. From the SEM micrographs, we could easily observe the difference that the porous 40wt%SSZ-SLT backbone was filled by GDC covering the SSZ-SLT particles surface.

The cross section of electrolyte/anode interface SEM micrograph was presented in Figure 4 (c). The EDX map for Figure 4 (c) was taken for the elements, Ce, Zr, and Ti, representing GDC, SSZ, and SLT respectively; the EDX maps of Ce, Zr, and Ti were shown in Figures 4 (d), 4 (e), and 4 (f), respectively. The EDX spectrum of Ce, Zr, and Ti was also shown in Figure 4 (g). The quantitative analysis for the mapping area indicated that the content of Ce, Zr, and Ti in atomic ratio was 39.4%, 29.7%, and 30.9% with an error margin of 2.4%, which was slightly different from respective atomic ratio, 31.3%, 32.8%, and 35.9%, calculated from the anode fabrication process. The difference would probably come from the local inhomogeneous GDC distribution during the impregnation process, and GDC slightly aggregated in the area presented in

Figure 4. SEM micrographs of (a) 40wt%SSZ-SLT backbone, (b) GDC-impregnated 40wt%SSZ-SLT backbone, (c) cross section of GDC-impregnated 40wt%SSZ-SLT anode functional layer; EDX maps of (d) Ce, (e) Zr, and (f) Ti in the anode functional layer; (g) EDX spectrum of Ce, Zr, and Ti in the anode functional layer.

the SEM micrograph of Figure 4 (c). It is surely important to analyze the dispersity of the impregnated catalyst should be evaluated, however, it is not easily accessible from current SEM-EDX resolution limit for such nano-scale three-dimensional internal structure of the anode. Further work should pay attentions to impregnation processing, and should analyze such nano-structure by using e.g. three-dimentional TEM tomography and high-resolution focused-ion-beam (FIB) electron microscopy.

3.2. Performance promotion by 0.03 mgcm^{-2} metal catalyst impregnation

In order to further improve cell performance, a secondary metal catalyst was further incorporated into the pure oxide anode layer; nevertheless, the loading amount of metal phase catalyst should be carefully controlled. According to our previous study, the existence of a large amount of metal catalyst will suffer serious agglomeration, and cause performance deterioration especially under a highly humidified condition and under the redox cycling conditions, while the oxide catalyst, ceria, demonstrated much better redox-resistance, since it has less tendency of agglomeration in such an operating condition [27, 28]. As a result, the anodes for both high performance and high

redox-resistance should have a high content of oxide catalyst and a low content of metal catalyst.

The additional 0.03 mgcm^{-2} metal catalyst was then incorporated, which was 1/500 of the GDC loading, 15 mgcm^{-2} , and it was around 0.07wt% of the ceramic phases. The impregnation loading of oxide catalyst and metal catalyst was thus considered as an extreme situation in our study. I-V curves of SOFC single cells using the GDC-impregnated 40wt%SSZ-SLT oxide anode and the Ni- or Pd-incorporated oxide

Figure 5. (a) I-V and I-P curve of SOFC single cell using the GDC/SSZ-SLT anode, the Ni-promoted GDC/SSZ-SLT anode, and the Pd-promoted GDC/SSZ-SLT anode; (b) I-V curve of SOFC single cell using the GDC/SSZ-SLT anode, the Ni-promoted GDC/SSZ-SLT anode, the Pd-promoted GDC/SSZ-SLT anode, and the Ni-SSZ anode in a low current density region up to 400 mAcm^{-2} .

anode were presented in Figure 5 (a). Cell performance was considerably improved by the impregnation with only a small amount of metal catalyst. Figure 5 (b) gives the comparison of the three different oxide-based anodes with the conventional Ni-SSZ anode in the low current density region where the fuel cells normally operate. With the additional 0.07wt% metal phase catalyst, single cell performance was improved from around 0.865V to 0.974V at 200 mAcm⁻². The presence of Pd or Ni nanoparticles may strongly promote the reduction of surface Ce⁴⁺ through hydrogen spillover mechanism [29, 30]; on the other hand, the oxygen chemisorption of Pd or Ni was also taken place by oxygen spillover that chemisorbed oxygen would come from the steam in the fuel and the oxygen ion passing through the GDC surface. The increase of concentration of O_{ads} or the formation of sub-surface oxide sites on the Pd or Ni surface further enhanced the hydrogen adsorption/dissociation and hydrogen diffusion processes on the metal surface [30, 31]. As a result, even a small amount of Pd or Ni located on highly-conductive doped-ceria surface could significantly lower the activation energy and promote hydrogen oxidation reaction. Other structure-related phenomenon was negligible since such low catalyst loading cannot change structural parameter of the anode.

3.3. Redox stability of the metal-incorporated oxide anode

Redox cycling test was performed to investigate redox stability of the anodes by using the fuel interrupt method applied previously [25]. In the present study, totally 50

Figure 6. Terminal voltage of single cell using (a) the Pd-promoted oxide anode, and (b) the Ni-promoted oxide anode during the first 10 redox cycles; (c) anode-side ohmic loss of single cell using the Ni-promoted oxide anode and the Pd-promoted oxide anode during 50 redox cycles; (d) anodic overvoltage of single cell using the Ni-promoted oxide anode and the Pd-promoted oxide anode during 50 redox cycles.

redox cycles were repeated during the cycling test. Figures 6 (a) and (b) show the terminal voltage of the SOFC single cell using the Pd- and Ni-promoted anodes during the first 10 redox cycles, respectively. No obvious difference could be detected from the first 10 cycles, and we further separated the anode-side ohmic loss and anodic overvoltage of both anodes against the entire 50 redox cycles in Figures 6 (c) and (d). The stable anode-side ohmic loss directly demonstrated the robustness of our anode backbone structure that SLT-SSZ provides sufficient electrical conductivity and shows negligible thermal and chemical expansion coefficient [9, 13]. Although doped ceria shows large chemical expansion [32], the impregnated nano-particles would not deteriorate the 1300°C sintered SLT-SSZ structure. On the other hand, anodic overvoltage of both anodes increased as the redox cycling test was performed, nevertheless, the degradation using the different metal catalysts was slightly different: the Ni-promoted oxide anode continuously degraded as the redox cycle was repeated, and its overvoltage loss at the constant current 200 mAcm^{-2} increased from 44 mV to 55 mV, and to 71 mV upon the 10th and 50th cycle, respectively. In the case of Pd-promoted oxide anode, the degradation within the first 10 redox cycles was much more serious, overvoltage increased from 44 mV to 58 mV, but it was stabilized after 10 redox cycles

with the overvoltage loss of 59 mV at the 50th redox cycle at the constant current density of 200 mAcm⁻².

I-V curve of SOFC single cells using the different anodes after the 50 redox cycles

Figure 7. I-V curve of SOFC single cell using (a) the Pd-promoted GDC/SSZ-SLT anode, and (b) the Ni-promoted GDC/SSZ-SLT anode before and after 50 redox cycles.

was measured again. Figure 7 (a) and (b) gives the comparison of I-V curves of single cells using the different anodes before and after the cycling test. The voltage difference gradually increased as the current density increased, nevertheless, in the low current density region as measured at 200 mAcm^{-2} , cell voltage dropped 2.9% (from 0.971V to 0.943V) when Ni was employed, but in the case of using Pd, cell voltage dropped 2.5% (from 0.973V to 0.949V). The degraded cells even achieved better performance than cells using the fresh Ni-SSZ cermet anode, cell voltage achieved $\sim 0.9\text{V}$ at 200 mAcm^{-2} in the same operating conditions, which has a degradation rate up to 7.5% per 10 redox cycles against the redox cycling test [25]. The degradation of the Ni-SSZ anode was systematically investigated in our previous study correlating to the change in anode-side ohmic loss and anodic overvoltage against different degradation patterns. We found that both ohmic loss and overvoltage of Ni-SSZ largely increased as the redox cycling test was performed, and concluded that the shrinkage and expansion of Ni during redox cycling deteriorated the electrical conducting path and reduced electrochemical reaction sites in the anode which caused the redox instability of the conventional Ni-SSZ cermet [25, 28]. The catalyst-impregnated anode was reported having improved redox stability; the ceramic anode backbone ensures the stable electrical conducting path showing no change in ohmic loss, and the homogeneously dispersed catalyst could minimize the

reaction site loss [20, 33-34]. However, we found that the catalyst species largely influenced the redox stability, since oxide catalyst, ceria, has much less tendency in aggregation than that of metal catalyst, Ni. The partial substitution of Ni by CeO₂ improved the redox stability but decreased the cell performance [28]. In the current study, both high performance and high redox stability could be achieved using 0.03 mgcm⁻² GDC as the main catalyst and 0.03 mgcm⁻² Ni or Pd as the co-catalyst with controlled catalyst loading.

3.4. Microstructure evolution upon the redox cycling test

The SEM micrographs of the degraded anodes were shown in Figures 8 (a)-(d) with carbon coating to ensure clear SEM observation. Figures 8 (a) and (c) and Figures 8 (b) and (d), respectively, show the Pd and Ni nano-particles located on the oxide surface with a diameter around 100 nm. However, it was difficult to detect a large amount of Pd and Ni particles in the limited surface area, since only 0.03 mgcm⁻² metal catalyst was incorporated into the anodes. Besides, it should be emphasized that the catalyst was impregnated into the anode layer and it contained in both the current conducting layer and the functional layer. As the result, the metal catalyst included in the functional layer

would be much less than 0.03 mgcm^{-2} . The identification of Pd and Ni was performed by EDX spectra point analysis shown in Figures 8 (e) and (f); the weak peaks at around 2.8 keV were identified as the energy spectra of Pd $L\alpha$, and the peaks at 7.5 keV and 8.2 keV were identified as the energy spectra of Ni $K\alpha$ and Ni $K\beta$, respectively.

The excellent redox stability of the metal catalyst promoted oxide anodes as

Figure 8. The microstructure of (a), (c) the Pd-promoted GDC/SSZ-SLT anode, and (b), (d) the Ni-promoted GDC/SSZ-SLT anode after 50 redox cycles; EDX spectra of (e) Pd nano-particles, and (f) Ni nano-particles.

described in this study should also be ascribed to the low concentration of impregnated metal catalysts. The low metal catalyst concentration prevents the serious particle aggregation upon the redox cycling test and suppresses the anodic overvoltage increase. The evolution of catalyst particles would be the agglomeration of closely adjacent particles from the initial impregnation stage. If not, serious metal particle aggregation would occur upon redox cycling test in impregnating 2.88 mgcm^{-2} Ni (10wt% of ceramic phase) as shown previously [28].

4. Conclusions

The stable 40wt%SSZ-SLT cermet structure was utilized as the backbone for the SOFC anode because of its high conductivity, identical thermal expansion coefficient, and negligible chemical expansion coefficient. The secondary Gd-doped CeO_2 conducting and catalytically-active phase was impregnated to coat the SSZ-SLT backbone surface. The GDC-coated SSZ-SLT pure oxide anode demonstrated comparable electrochemical performance with the conventional Ni-SSZ anode in 3%-humidified hydrogen fuel at 800°C. In order to further improve electrochemical performance of the anode, additional 0.03 mgcm^{-2} metal catalysts, Ni or Pd, were

incorporated into the anode by impregnation. The extremely low concentration metal catalyst substantially improved electrochemical properties and cell performance with excellent redox stability, and the deteriorated cells even retained higher performance than the fresh cell using the conventional Ni-SSZ. As the consequence, the GDC-coated 40wt%SSZ-SLT oxide with the Ni or Pd promotion can be an alternative SOFC anode with both high performance and excellent redox stability.

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