

Electrochemical Activity of NiCo-GDC Cermet Anode for Solid Oxide Fuel Cells Operated in Methane

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Abstract : Solid Oxide Fuel Cells (SOFCs) have been considered as one of the most efficient large unit power generators for household and industrial applications. The efficiency of an electronic cell depends mainly on the electrochemical reactions in the anode. The development of anode materials has been intensely studied to achieve higher kinetic rates of redox reactions and lower internal resistance. Recent studies have introduced an efficient cermet (ceramic-metallic) material for its ability in fuel oxidation and oxide conduction. This could expand the reactive site, also known as the triple-phase boundary (TPB), thus increasing the overall performance. In this study, a bimetallic catalyst $\text{Ni}_{0.75}\text{Co}_{0.25}\text{O}_x$ was combined with $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{1.95}$ (GDC) to be used as a cermet anode (NiCo-GDC) for an anode-supported type SOFC. The synthesis of $\text{Ni}_{0.75}\text{Co}_{0.25}\text{O}_x$ was carried out by ball milling NiO and Co_3O_4 powders in ethanol and calcined at 1000 °C. The $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{1.95}$ was prepared by a urea coprecipitation method. Precursors of $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ and $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ were dissolved in distilled water with the addition of urea and were heated subsequently. The heated mixture product was filtered and rinsed thoroughly, then dried and calcined at 800 °C and 1500 °C, respectively. The two powders were combined followed by pelletization and sintering at 1100 °C to form an anode support layer. The fabrications of an electrolyte layer and cathode layer were conducted. The electrochemical performance in H_2 was measured from 800 °C to 600 °C while for CH_4 was from 750 °C to 600 °C. The maximum power density at 750 °C in H_2 was 13% higher than in CH_4 . The difference in performance was due to higher polarization resistances confirmed by the impedance spectra. According to the standard enthalpy, the dissociation energy of C-H bonds in CH_4 is slightly higher than the H-H bond H_2 . The dissociation of CH_4 could be the cause of resistance within the anode material. The results from lower temperatures showed a descending trend of power density in relevance to the increased polarization resistance. This was due to lowering conductivity when the temperature decreases. The long-term stability was measured at 750 °C in CH_4 monitoring at 12-hour intervals. The maximum power density tends to increase gradually with time while the resistances were maintained. This suggests the enhanced stability from charge transfer activities in doped ceria due to the transition of $\text{Ce}^{4+} \leftrightarrow \text{Ce}^{3+}$ at low oxygen partial pressure and high-temperature atmosphere. However, the power density started to drop after 60 h, and the cell potential also dropped from 0.3249 V to 0.2850 V. These phenomena was confirmed by a shifted impedance spectra indicating a higher ohmic resistance. The observation by FESEM and EDX-mapping suggests the degradation due to mass transport of ions in the electrolyte while the anode microstructure was still maintained. In summary, the electrochemical test and stability test for 60 h was achieved by NiCo-GDC cermet anode. Coke deposition was not detected after operation in CH_4 , hence this confirms the superior properties of the bimetallic cermet anode over typical Ni-GDC.

Keywords : bimetallic catalyst, ceria-based SOFCs, methane oxidation, solid oxide fuel cell

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