



Feasibility of simultaneous NO reduction and electricity generation in SOFCs with V₂O₅ or Cu added LSCF-GDC cathodes

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ARTICLE INFO

Article history:

Received 25 November 2008
Received in revised form 8 December 2008
Accepted 9 December 2008
Available online 16 December 2008

Keywords:

Nitric oxide
Electrochemical reduction
Solid oxide fuel cell
Oxygen concentration
Maximum power density
Synergistic effect

ABSTRACT

Solid oxide fuel cell (SOFC) units were constructed with Ni-GDC (gadolinia-doped ceria) as the anode, YSZ (yttria-stabilized zirconia) as the electrolyte, and V₂O₅ or Cu added La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF)-GDC composites as the cathodes, respectively. The electrochemical NO reduction occurs over either V₂O₅ or Cu added LSCF-GDC cathode without or with the presence of oxygen. The maximum power density decreases with decreasing O₂ concentration. When the O₂ concentration is 3% or larger, simultaneous NO reduction and electricity generation in the SOFCs can be not only feasible but also beneficial via a synergistic effect.

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1. Introduction

The removal of nitric oxide (NO) from industrial flue gas in stationary sources is traditionally performed by the selective catalytic reduction (SCR) process, in which NO in the flue gas reacts with ammonia to produce nitrogen and water [1]. This SCR process has to use a reducing gas, usually ammonia, while residual ammonia needs additional treatment. Thus, the electrochemical NO reduction without a reducing gas has been studied extensively [2–5]. However, this process of electrochemical NO reduction is performed with applied current and the current efficiency is generally only a few percent; additionally, simultaneous O₂ reduction occurs to consume substantial amount of electrical current [5]. Therefore, simultaneous NO reduction and electricity generation in a solid oxide fuel cell (SOFC) should be an attractive process; nevertheless, as far as the authors know, such a process has not yet been reported.

V₂O₅ is the catalytic component of the commercial SCR catalyst [6] and Cu is effective for direct electrochemical reduction of nitric oxide [5]. In this work, these catalytic components were added into the cathode materials of La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF)-gadolinia-doped ceria (GDC) composite, which exhibits good cathode performance [7]. LSCF is the well-known material for the cathode in intermediate temperature SOFCs, and adding GDC into LSCF to make the LSCF-GDC composite as the cathode can increase the

oxygen reduction activity of the cathode [7]. Additionally, a process of simultaneous NO reduction and electricity generation means that the SOFC operation is performed with the flue gas passing the cathode-side; thus, the O₂ concentration is much smaller than the normal 20%. For the flue gas of the power plant or the nitric acid plant, the O₂ concentration is usually 4–6% and the NO concentration can be 1000–5000 ppm or larger. In this work, the effects of the O₂ concentrations on the SOFC performance without or with NO were investigated. The results showed that simultaneous NO reduction and electricity generation in SOFCs can be not only feasible but also beneficial via a synergistic effect.

2. Experimental

2.1. Preparation of V₂O₅ and Cu added LSCF-GDC composites

LSCF was prepared by glycine-nitrate process. Appropriate amounts of reagent-grade (Showa, Japan) metal nitrates La(NO₃)₃·6H₂O, Sr(NO₃)₂, Co(NO₃)₂·6H₂O and Fe(NO₃)₃·9H₂O were dissolved in de-ionized water. Glycine (Sigma, USA) was also dissolved in de-ionized water. Then, these two solutions were mixed with glycine to NO₃ ratio of 1:0.8. The mixture was heated under stirring at 110 °C until combustion occurred. The product was ground to powders and then calcined by heating to 500 °C and held for 2 h, then to 900 °C and held for 4 h. The heating was always done in air at 5 °C/min. LSCF is La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ}.

GDC was prepared by co-precipitation. The details of the method have been presented elsewhere [8]. The GDC powders were

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calcined by heating to 500 °C and held for 2 h, and then to 1000 °C and held for 2 h before cooling. GDC is (GdO_{1.5})_{0.1}(CeO₂)_{0.9}.

The LSCF-GDC composite was prepared by mixing the above-prepared LSCF and GDC powders at LSCF:GDC = 100:50 in weight. The mixture was ground for 24 h, then calcined by heating to 500 °C and held for 2 h, and then to 900 °C, held for 10 h.

LSCF-GDC-V₂O₅ was prepared by mixing V₂O₅ powder (Showa, Japan) with LSCF-GDC powder in de-ionized water. Cu adding to LSCF-GDC was done by impregnation with Cu(NO₃)₂ · 3H₂O (Showa, Japan) solution. After drying, the powders were calcined by heating to 500 °C and held for 2 h, and then to 800 °C and held for 2 h. The V₂O₅ and Cu loading of LSCF-GDC-V₂O₅ and LSCF-GDC-Cu, respectively, was 2 wt.% with respect to LSCF.

2.2. Current-voltage measurement in SOFC unit cell

A disk was cut from yttria-stabilized zirconia (YSZ) tape (156 μm thickness, Jiuhow, Taiwan) to make an electrolyte-supported cell. One side of the disk was spin-coated with Ni-GDC paste, with Ni:GDC = 60:100 in weight [9]. The other side of the disk was spin-coated with LSCF-GDC-V₂O₅ or LSCF-GDC-Cu powders to make the cathode layer. The details of the construction of the SOFC unit cell have been presented elsewhere [9]. The SOFC cell has an anode area of 1 cm², an anode thickness of 30 μm, an electrolyte thickness of 156 μm, a cathode area of 1 cm², and a cathode thickness of 10 μm. These thicknesses were measured from a scanning electron micrograph of the cross section of the cell.

The current-voltage measurements were performed at 800 °C with pure hydrogen flowing on the anode side. The cathode-side gas was various oxygen or oxygen plus NO mixture, all balanced by argon. The flow rate was always 150 cm³ min⁻¹. With the same cathode materials, the measurements were all performed on the same SOFC unit cell.

3. Results and discussion

3.1. V₂O₅ added LSCF-GDC cathode

Fig. 1 shows the variation of voltage-current and power-current profiles with NO concentration over either V₂O₅ or Cu added LSCF-GDC cathode. Fig. 1a shows that, with 2–6% NO over the LSCF-GDC-V₂O₅ cathode, the maximum power density increase with increasing NO concentration. The generation of power density or electrical current means the formation of the O species over the cathode. Since NO is the only oxygen source in this case, NO reduction or decomposition to produce the O species should have occurred. This indicates that direct electrochemical NO reduction can occur over the LSCF-GDC-V₂O₅ cathode when oxygen is not present. The direct electrochemical NO reduction with applied current can be considered to proceed via the following reaction [5]



where h⁺ is an electron hole. Thus, the O species during the NO reduction with simultaneous electricity generation may be O²⁻, the oxygen ion; nevertheless, what kind of O species is formed during simultaneous NO reduction and electricity generation in SOFCs needs further studies. However, the flue gas contains oxygen and thus NO treatment should be studied in the presence of oxygen. Additionally, since NO concentration affects the SOFC performance in terms of the maximum power density, the effect of the O₂ concentration on the SOFC performance should also be studied.

Table 1 shows that the maximum power density decreases with decreasing O₂ concentration; however, the maximum power density does not drop too much when the O₂ concentration decreases from 20% to 5%, the latter usually being the mid O₂ concentration

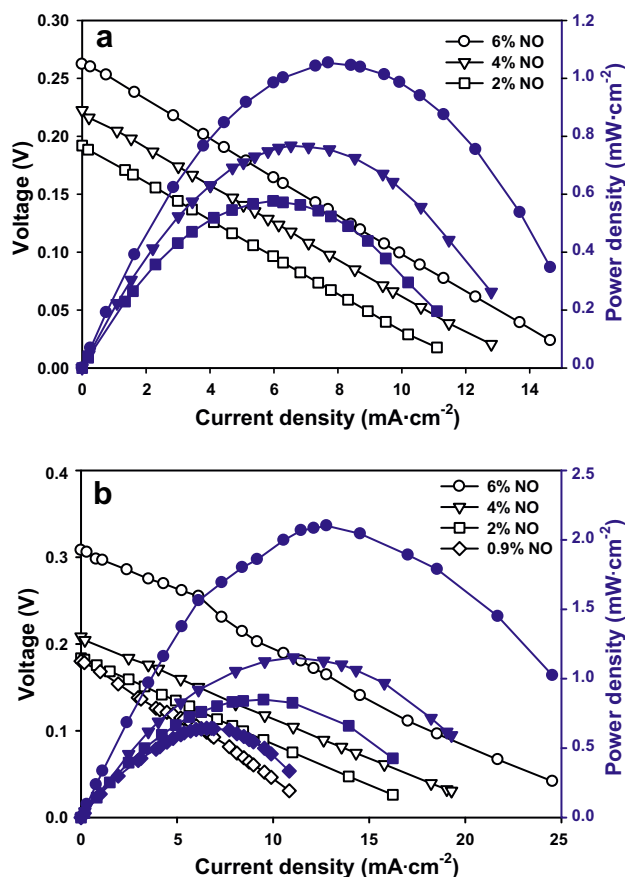


Fig. 1. Variation of voltage-current and power-current profiles with NO concentration over (a) V₂O₅ added LSCF-GDC cathode, and (b) Cu added LSCF-GDC cathode. Open symbol: voltage; filled symbol: power density.

Table 1

Variation of maximum power density with O₂ and NO concentrations over V₂O₅ added LSCF-GDC cathode.

Cathode gas	Maximum power density (mW cm ⁻²)
20% O ₂	38.99
10% O ₂	36.53
6% O ₂	31.82
6% O ₂ + 5000 ppm NO	32.15
6% O ₂ + 3000 ppm NO	32.01
6% O ₂ + 1000 ppm NO	31.82
5% O ₂	27.62
5% O ₂ + 5000 ppm NO	27.99
5% O ₂ + 3000 ppm NO	27.83
5% O ₂ + 1000 ppm NO	27.63
4% O ₂	26.72
4% O ₂ + 5000 ppm NO	27.12
4% O ₂ + 3000 ppm NO	26.94
4% O ₂ + 1000 ppm NO	26.74
3% O ₂	25.15
3% O ₂ + 5000 ppm NO	25.66
3% O ₂ + 3000 ppm NO	25.38
3% O ₂ + 1000 ppm NO	25.17
2% O ₂	22.73
2% O ₂ + 5000 ppm NO	23.81
2% O ₂ + 3000 ppm NO	23.54
2% O ₂ + 1000 ppm NO	22.84
1% O ₂	16.68
1% O ₂ + 5000 ppm NO	18.12
1% O ₂ + 3000 ppm NO	17.63
1% O ₂ + 1000 ppm NO	17.49

in the flue gas. Thus, simultaneous NO reduction and electricity generation would be feasible in terms of the SOFC performance.

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