



# Nitric oxide removal from simulated lean-burn engine exhaust using a solid oxide fuel cell with V-added (LaSr)MnO<sub>3</sub> cathode

Ta-Jen Huang\*, I.-Chuan Hsiao

Department of Chemical Engineering, National Tsing Hua University, 101 Kuang Fu Road, Sec. 2, Hsinchu 30013, Taiwan, ROC

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## ABSTRACT

A solid oxide fuel cell (SOFC) unit cell is constructed by coating V-added La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3</sub>-Ce<sub>0.9</sub>Gd<sub>0.1</sub>O<sub>2</sub> as the cathode onto a disk of anode-supported bilayers of yttria-stabilized zirconia (YSZ) and Ni-YSZ. Various compositions of lean-burn engine exhaust components of O<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub> and NO were tested over the cathode of SOFC unit cell operating at 700–800 °C. NO can be completely converted to N<sub>2</sub>. All the exhaust components have beneficial effect on NO conversion. Increasing O<sub>2</sub> content from 2 to 10% increases NO conversion from 18% to about 70%; this enables us to increase the O<sub>2</sub> content by adding air in the exhaust to increase the NO conversion. Increasing NO concentration increases NO conversion. Adding H<sub>2</sub>O, increasing CO<sub>2</sub> content and an increase of the generated voltage all increase NO conversion, although only slightly. These are beneficial for the application of SOFCs to NO removal from lean-burn engine exhaust.

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## 1. Introduction

The removal of nitric oxide (NO) from the lean-burn exhaust, such as the diesel engine exhaust, can be performed by the selective catalytic reduction (SCR) process, in which NO in the exhaust gases reacts with ammonia to produce nitrogen and water [1]. This SCR process has to use a reducing gas, usually ammonia; however, the residual ammonia can pose a problem. The technology of NO<sub>x</sub> storage-reduction (NSR) has also been considered for NO<sub>x</sub> removal. However, the NSR system needs to oxidize NO to NO<sub>2</sub> for storage and to periodically run the engine rich of stoichiometry for reduction [2]; this adds the complexity. Thus, the electrochemical NO reduction without a reducing gas has been studied extensively [3–8]. However, this process of electrochemical NO reduction is performed with applied current and the current efficiency is generally only a few percent because the reduction of O<sub>2</sub> occurs simultaneously to consume a substantial amount of the electrical current [8]. On the other hand, electrochemical NO reduction using a solid oxide fuel cell (SOFC) can remove NO and generate an electrical current instead [9–12]. Although this process needs an anode fuel, this fuel is consumed only when an electrical current is generated; thus, this SOFC-DeNO<sub>x</sub> process is very energy-efficient.

A process of DeNO<sub>x</sub> by SOFC means that the SOFC operation is performed with the exhaust gases passing the cathode side. For

the exhaust gases of the power plant or the nitric acid plant, the O<sub>2</sub> concentration is usually 4–6% and the NO concentration can be 1000–5000 ppm or higher. Recently, the problem of CO<sub>2</sub> emission has become very critical. The lean-burn engines are promising to decrease the fuel consumption for automotive use and thus decrease the CO<sub>2</sub> emission. However, for NO removal from the exhaust of the lean-burn engines, an efficient NO reduction at high oxygen concentrations is needed. Although the SOFC-DeNO<sub>x</sub> process can remove NO under high O<sub>2</sub> concentration, the reported experimental results until now have not shown its efficiency in the presence of water vapor; instead, it has been shown that the presence of 0.3% H<sub>2</sub>O in a simulated exhaust has inhibited the NO activity considerably [11]. Notably, the lean-burn engine exhaust contains about 10% H<sub>2</sub>O [13]. Therefore, suitable cathode materials in association with proper operating conditions are still to be studied for the SOFC-DeNO<sub>x</sub> process to become practical in automotive applications.

For the application of the SOFC-DeNO<sub>x</sub> process onto automobiles, the heating of SOFCs and thermal cycling may be problems; these may be solved using microtubular SOFCs with heating at the center of the tube that is on the anode side [14]. For microtubular SOFCs, quick heating can result in a startup time less than 10 s and thousands of thermal cycles can be operated [15]. This may be due to the mechanism that SOFC can become operational when only the electrolyte part of SOFC is heated to a temperature for startup of oxygen migration. Notably, SOFC is well known to produce more heat than needed to sustain its operating temperature. Additionally, the supply of the anode fuel may also be a problem; this has

\* Corresponding author. Tel.: +886 3 5716260; fax: +886 3 5715408.  
E-mail address: [tjhuang@che.nthu.edu.tw](mailto:tjhuang@che.nthu.edu.tw) (T.-J. Huang).

been solved by the advance in the fuel processing technology, for example, SOFC-based auxiliary power unit for diesel trucks can use diesel as the fuel [16].

(LaSr)MnO<sub>3</sub> (LSM) perovskite is the commercial cathode material for SOFCs. The addition of gadolinia-doped ceria (GDC) into the perovskites can increase the oxygen reduction activity of the cathode [17]. Preliminary test in this work has shown that LSM-GDC is effective for NO conversion in the presence of 10% H<sub>2</sub>O to allow us to simulate the lean-burn engine exhaust. Thus, the LSM-GDC composite is the choice of SOFC cathode material in this work. Additionally, V<sub>2</sub>O<sub>5</sub> is the catalytic component of the commercial SCR catalyst [18]. The addition of this catalytic component into LSM is considered to be able to enhance the NO activity.

In this work, the current–voltage measurement and the fixed-voltage test were performed in an SOFC unit cell with V-added LSM-GDC cathode. Various compositions of lean-burn engine exhaust components of O<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub> and NO were tested over the SOFC cathode at 700–800 °C. All these exhaust components have shown positive effect on NO conversion. This is beneficial for the application of the SOFC-DeNO<sub>x</sub> process to NO removal from the lean-burn engine exhaust.

## 2. Experimental

### 2.1. Preparation of LSM and GDC and V addition

LSM was prepared by the glycine-nitrate process with a composition of La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3</sub>. Appropriate amounts of reagent-grade (Showa, Japan) metal nitrates La(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O, Sr(NO<sub>3</sub>)<sub>2</sub>, and Mn(NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O were dissolved in de-ionized water. Glycine (Sigma, USA) was also dissolved in de-ionized water. Then, these two solutions were mixed together with a glycine to NO<sub>3</sub><sup>-</sup> ratio of 0.8:1. The mixture was then heated under stirring at 110 °C until combustion occurred. The product was ground to powders. Then, the powders were calcined by heating to 500 °C and held for 2 h, then to 900 °C and held for 4 h, and then slowly cooled down to room temperature. In this work, the heating of the powders was always done in air at a rate of 5 °C min<sup>-1</sup>.

GDC was prepared by the co-precipitation method with a composition of Ce<sub>0.9</sub>Gd<sub>0.1</sub>O<sub>2</sub>. The details of the method have been described elsewhere [19]. The GDC powders were calcined by heating to 900 °C and held for 4 h.

The LSM-GDC composite was prepared by mixing the above-prepared LSM and GDC powders at a ratio of LSM:GDC being 1:1 in weight. The mixture was ground for 24 h, then calcined by heating to 500 °C and held for 2 h, and then to 800 °C, held for 4 h.

Adding V to LSM-GDC powders was done by impregnation. The V cation solution was prepared by dissolving NH<sub>4</sub>VO<sub>3</sub> (Merck, Germany) in de-ionized water. After drying, the powders were calcined by heating to 800 °C and held for 4 h before cooling down to room temperature. Notably, after calcination at 800 °C when O<sub>2</sub> can be dissociated to oxygen atom over V added LSM-GDC, V can be fully oxidized to V<sub>2</sub>O<sub>5</sub> [20]. The weight of V<sub>2</sub>O<sub>5</sub> in calcined V-added LSM-GDC was 1 wt.% in terms of the weight of LSM.

### 2.2. Construction of SOFC unit cell

A commercial disk of anode-supported bilayers of yttria-stabilized zirconia (YSZ) and Ni-YSZ (NexTech, USA) was used to make an anode-supported cell. The disk has a diameter of 2 cm. The cathode side of the disk was spin-coated with a thin interlayer of LSM-GDC to enhance adhesion; the thus-coated disk was dried at 50 °C for 6 h, then heated in air at 10 °C min<sup>-1</sup> to 500 °C and held for 2 h, and then heated at 5 °C min<sup>-1</sup> to 1200 °C and held for 2 h before cooling down. Then, the disk was spin-coated with V-added

LSM-GDC as the cathode functional layer; the thus-coated disk was dried at 50 °C for 6 h, then heated at 10 °C min<sup>-1</sup> to 500 °C and held for 2 h, and then heated at 5 °C min<sup>-1</sup> to 900 °C and held for 2 h. Then, the disk was spin-coated with LSM as the current collecting layer to complete the SOFC unit cell; the heat treatment was the same as that for the cathode functional layer.

### 2.3. Current–voltage measurement

The measurement of current–voltage curve was performed at 700–800 °C with pure hydrogen flowing on the anode side and various gas mixtures, all balanced by helium, on the cathode side. The overall flow rate was 150 Nml/min and that on the anode side was the same as that on the cathode side. The voltage was varied by an adjustable resistor, and both the voltage and the current were measured by a Multimeter (TES 2730).

### 2.4. Fixed-voltage test

The fixed-voltage (constant-voltage) tests were performed at 800 °C with the generated voltage kept constant at 0.6 V, except noted otherwise. The anode gas was pure hydrogen. The inlet cathode gas was a mixture of NO, O<sub>2</sub>, H<sub>2</sub>O and/or CO<sub>2</sub>, balanced in helium, with the composition designated in the figure and table legends. The overall flow rate was always 150 Nml/min.

The tests were conducted with introducing a designated gas mixture to the cathode side of the SOFC unit cell until a steady state was obtained for a certain period of time. Throughout the test, the electrical current, the voltage, and the outlet gas compositions were always measured. The NO and NO<sub>2</sub> contents in the outlet cathode gas were measured by NO and NO<sub>2</sub> analyzers (NGA 2000, Emerson, Germany), respectively. The N<sub>2</sub> content was measured by a gas chromatograph (GC) equipped with a thermal conductivity detector (China Chromatography 8900, Taiwan).

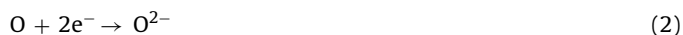
## 3. Results and discussion

### 3.1. Effect of O<sub>2</sub> concentration on NO conversion

Fig. 1 shows that an electrical current is generated with 6% NO as the cathode gas at 700–800 °C. This indicates the occurrence of electrochemical NO reduction—that is, the occurrence of NO decomposition:



followed by the charge transfer reaction to form the oxygen ion:



Notably, electrochemical NO reduction is completed and generates an electrical current only after the oxygen ion produced in reaction (2) has been transported to the anode and consumed by electrochemical oxidation with the fuel.

Table 1 shows that, with 6% NO as the cathode gas, the maximum power density increases with increasing temperature from 700 to 800 °C, being a same trend as that with 20% O<sub>2</sub>; however, the open circuit voltage (OCV) decreases with increasing temperature; this is also a same trend as that with 20% O<sub>2</sub> but at a much larger extent. Notably, the OCV with 20% O<sub>2</sub> as the cathode gas is close to the theoretical value, indicating that the SOFC setup of this work operated properly—that is, no gas leakage or cross over the electrolyte layer.

Table 2 shows a comparison of SOFC performance with various O<sub>2</sub> content in simulated exhaust gas containing 10% H<sub>2</sub>O, 10% CO<sub>2</sub> and 5000 ppm NO and that in a gas mixture containing only O<sub>2</sub> in He. Both the OCV and the maximum power density differ only

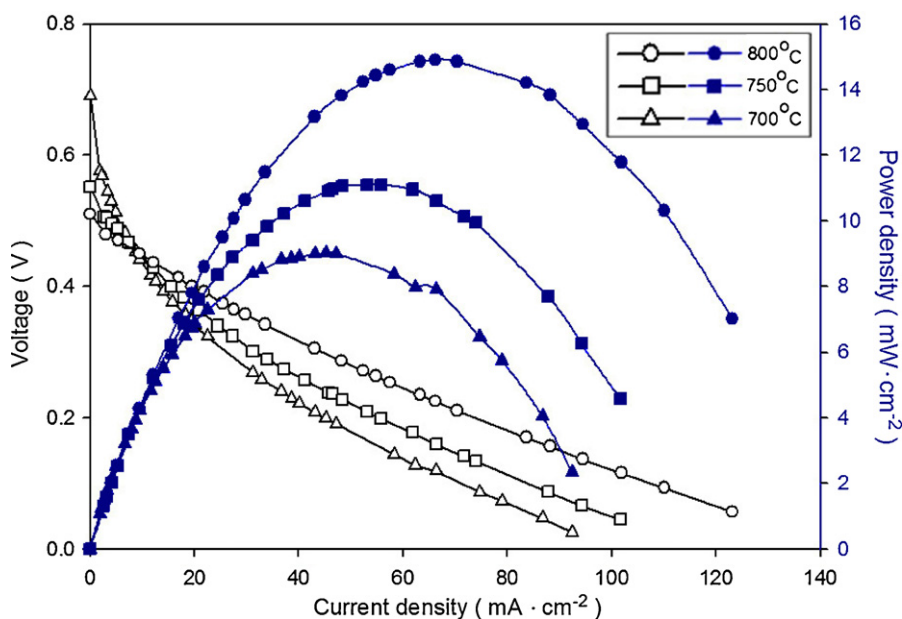


Fig. 1. Voltage–current and power–current profiles with 6% NO fed to the cathode. Empty symbol: voltage; filled symbol: power density.

Table 1

Effect of temperature on open circuit voltage and maximum power density with 6% NO and 20% O<sub>2</sub>, respectively, fed to the cathode.

Temperature (°C)	Open circuit voltage (V)	Maximum power density <sup>a</sup> (mW cm <sup>-2</sup> )
6% NO		
700	0.69	9.0
750	0.55	11.1
800	0.51	14.9
20% O <sub>2</sub>		
700	1.14	64.6
750	1.12	92.0
800	1.12	119.5

<sup>a</sup> Obtained via data from the current–voltage measurements.

slightly between these gas mixtures when the O<sub>2</sub> content varies from 2 to 10%. Notably, the major exhaust gas components are O<sub>2</sub>, water vapor (H<sub>2</sub>O) and CO<sub>2</sub>. On the other hand, the OCV increases slightly but the maximum power density does considerably with increasing O<sub>2</sub> content.

Fig. 2A and B shows the the steady state profiles of outlet NO concentrations with inlet NO concentrations of 600 and 5000 ppm, respectively, in simulated exhaust containing 10% H<sub>2</sub>O, 10% CO<sub>2</sub> and various O<sub>2</sub> content. Notably, the vertical lines between these steady states as shown in Fig. 2 and similar figures latter are drawn by the drawing software and do not indicate any actual behav-

Table 2

Effect of O<sub>2</sub> concentration on open circuit voltage and maximum power density at 800 °C.

O <sub>2</sub> concentration (%)	Simulated exhaust gas <sup>a</sup>		Gas mixture of O <sub>2</sub> in He	
	Open circuit voltage (V)	Maximum power density (mW cm <sup>-2</sup> )	Open circuit voltage (V)	Maximum power density (mW cm <sup>-2</sup> )
2	1.05	60.6	1.02	59.7
4	1.06	76.9	1.08	87.2
6	1.09	93.7	1.09	94.8
8	1.10	101.7	1.10	98.5
10	1.11	105.9	1.10	104.4

<sup>a</sup> With 10% H<sub>2</sub>O, 10% CO<sub>2</sub> and 5000 ppm NO in He.

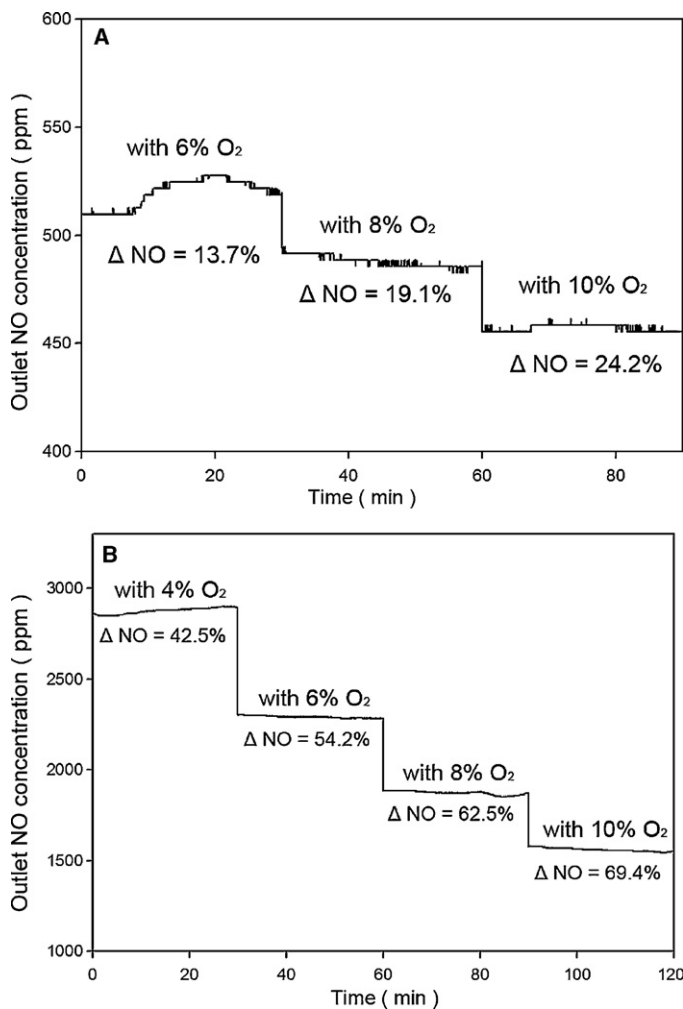


Fig. 2. Effect of O<sub>2</sub> concentration on NO conversion ( $\Delta$ NO) during steady state in fixed-voltage test at 800 °C with cathode gases of 10% H<sub>2</sub>O, 10% CO<sub>2</sub> and (A) 600 ppm NO, (B) 5000 ppm NO.

ior. The NO conversion increases considerably with increasing O<sub>2</sub> content, reaching about 70% at 10% O<sub>2</sub> with 5000 ppm NO in the exhaust. This is beneficial for NO removal from the exhaust during the aftertreatment since air can be added to increase the O<sub>2</sub> content. This is also beneficial for power generation of the SOFCs since increased O<sub>2</sub> content also increases the power density as shown in Table 2.

The enhancement of NO conversion by increased O<sub>2</sub> content is attributed to a synergism between NO reduction and O<sub>2</sub> reduction. An explanation for this synergism may be that O<sub>2</sub> reduction increases the concentration of the O species over the cathode and thus the generated potential (voltage) increases [21] and vice versa; then, increasing potential increases NO conversion as will be shown in Section 3.4. Notably, electrochemical NO reduction occurs according to reactions (1) and (2). Similarly, electrochemical O<sub>2</sub> reduction occurs also first with O<sub>2</sub> decomposition:



followed by the charge transfer reaction (2). Notably, also, there is no discrimination in the source of the O species for the occurrence of reaction (2). Therefore, when the O<sub>2</sub> concentration increases, a larger amount of the O species is formed and thus the rate of reaction (2) increases; this can increase the rate of NO decomposition reaction (1). This increase of the rate of NO decomposition is due to the transformation of larger amount of the formed O species to oxygen ions. Notably, also, the oxygen ions can be incorporated into the oxygen vacancies of LSM-GDC. Therefore, the NO conversion increases.

On the other hand, Fig. 2 shows that the NO conversion with 600 ppm NO is much smaller than that with 5000 ppm NO. This is attributed to smaller synergism between NO reduction and O<sub>2</sub> reduction due to lower NO concentration. Notably, this low NO concentration of 600 ppm is usually that in the lean-burn engine exhaust [13]. Thus, for the treatment of exhaust gases with low NO concentration, further studies are needed to find cathode materials that can increase the NO conversion at this low NO concentration. Notably, when Cu-added La<sub>0.58</sub>Sr<sub>0.4</sub>Co<sub>0.2</sub>Fe<sub>0.8</sub>O<sub>3</sub> is used as the cathode material for electrochemical NO<sub>x</sub> reduction in an SOFC operating at 800 °C, the NO conversion increases with decreasing NO concentration from 5000 to 150 ppm and 100% NO conversion is attained for inlet NO concentration of 150 ppm; however, the NO activity is inhibited considerably in the presence of 0.3% H<sub>2</sub>O [10]. Additionally, when Ag/La<sub>0.6</sub>Ce<sub>0.4</sub>CoO<sub>3</sub> catalyst is used for direct NO decomposition in the presence of 8% O<sub>2</sub> at 400 °C, the NO conversion also increases with decreasing NO concentration from 2000 to 200 ppm and over 70% NO conversion is attained for NO concentration smaller than 500 ppm; however, the presence of 0.5% CO<sub>2</sub> decreases the NO conversion considerably at 450 °C [22]. These reports show the possibility to attain relatively high NO conversion at low NO concentration; however, they also show that it is important to study the effects of water vapor and CO<sub>2</sub> on NO conversion.

### 3.2. Effect of water vapor on NO conversion

Fig. 3A and B shows the steady state profiles of outlet NO concentrations with O<sub>2</sub> content of 2 and 10%, respectively, in simulated exhaust containing 10% CO<sub>2</sub> and 5000 ppm NO with or without the presence of H<sub>2</sub>O. The presence of 10% H<sub>2</sub>O increases the NO conversion over that without H<sub>2</sub>O, although only slightly. We also observed that the maximum power density with the presence of H<sub>2</sub>O also differs only slightly from that without the presence of H<sub>2</sub>O, being 106 mW cm<sup>-2</sup> when no H<sub>2</sub>O was present in the simulated exhaust containing 10% O<sub>2</sub>, 10% CO<sub>2</sub> and 5000 ppm NO in comparison to that of 105.9 mW cm<sup>-2</sup> with 10% H<sub>2</sub>O as shown in Table 2.

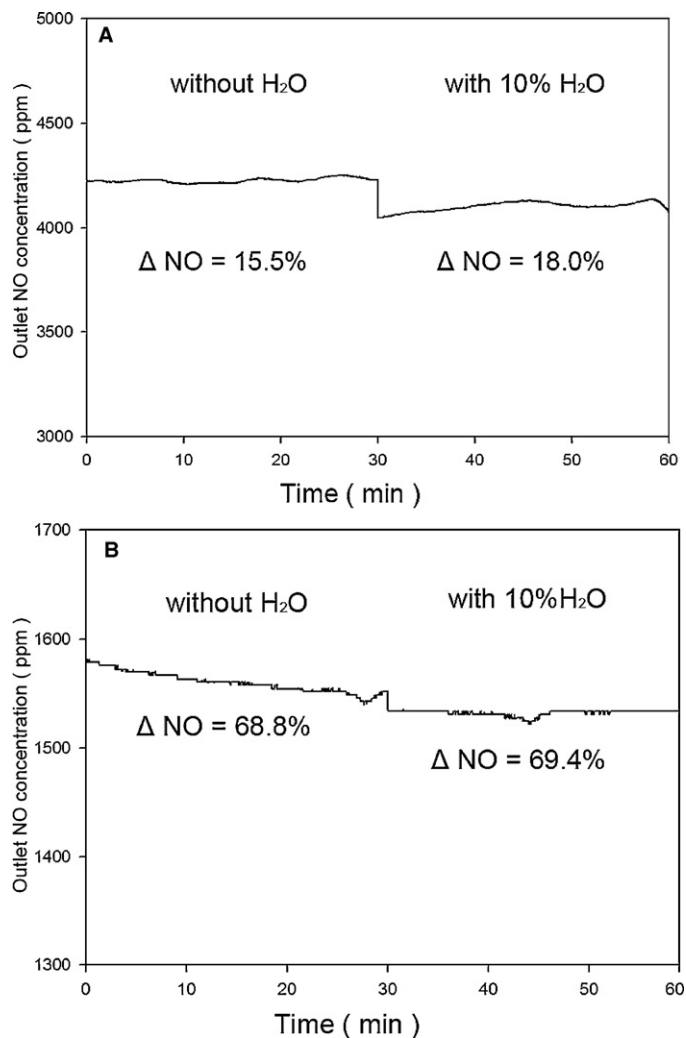


Fig. 3. Effect of H<sub>2</sub>O on NO conversion ( $\Delta\text{NO}$ ) during steady state in fixed-voltage test at 800 °C with cathode gases of 10% CO<sub>2</sub>, 5000 ppm NO and (A) 2% O<sub>2</sub>, (B) 10% O<sub>2</sub>.

The increase of the NO conversion in the presence of H<sub>2</sub>O can be attributed to the formation of H<sub>2</sub> from H<sub>2</sub>O dissociation [23]:



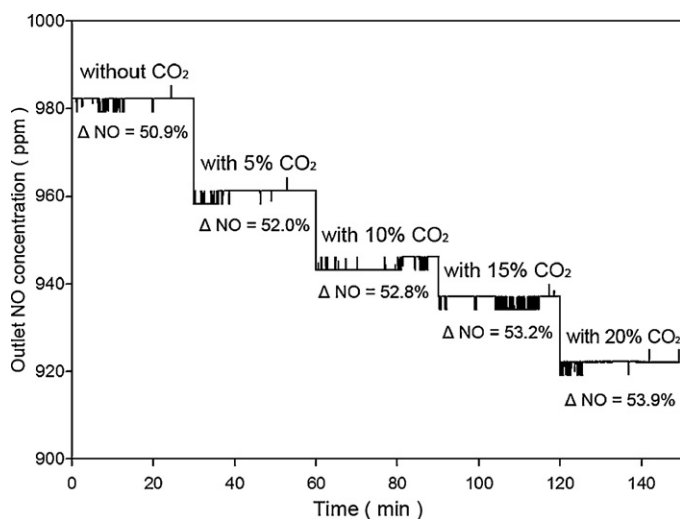
with the formed O species also incorporated into the oxygen vacancy after the charge transfer reaction (2). The formed H<sub>2</sub> should be able to reduce some NO and thus the NO conversion can increase. However, under environment of high temperature and high O<sub>2</sub> concentration such as that in this work, most H<sub>2</sub> should be oxidized via the reverse of reaction (4). Restated, H<sub>2</sub>O dissociation should be inhibited under high O<sub>2</sub> concentration.

### 3.3. Effect of CO<sub>2</sub> concentration on NO conversion

Fig. 4 shows that the NO conversion increases with increasing CO<sub>2</sub> content from 0 to 20%. This can be attributed to the formation of CO over the cathode; notably, CO<sub>2</sub> dissociation can occur over the cathode to produce CO [24]:



Notably, CO is a reducing agent for NO reduction [5,8]; thus, the NO conversion can increase. However, under environment of high temperature and high O<sub>2</sub> concentration such as that in this work, most CO should be oxidized via the reverse of reaction (5). Restated,



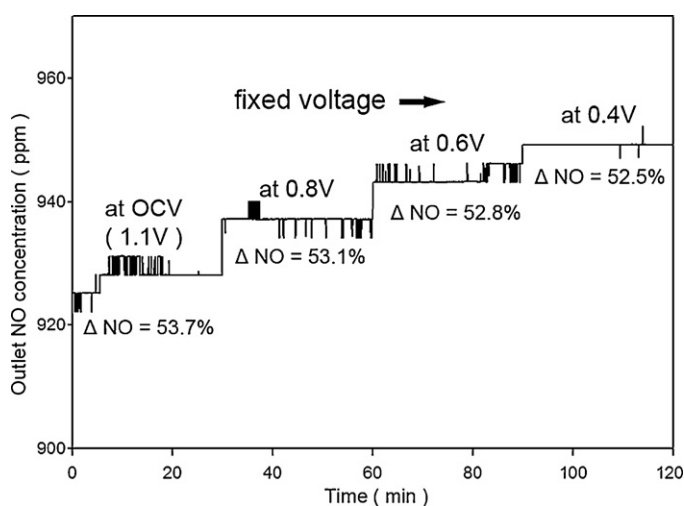
**Fig. 4.** Effect of CO<sub>2</sub> concentration on NO conversion ( $\Delta$ NO) during steady state in fixed-voltage test at 800 °C with cathode gases of 10% O<sub>2</sub>, 10% H<sub>2</sub>O and 2000 ppm NO.

CO<sub>2</sub> dissociation should be inhibited under high O<sub>2</sub> concentration. Nevertheless, very small amount of CO was indeed observed in the gas product when only CO<sub>2</sub> and O<sub>2</sub> were present in the feed.

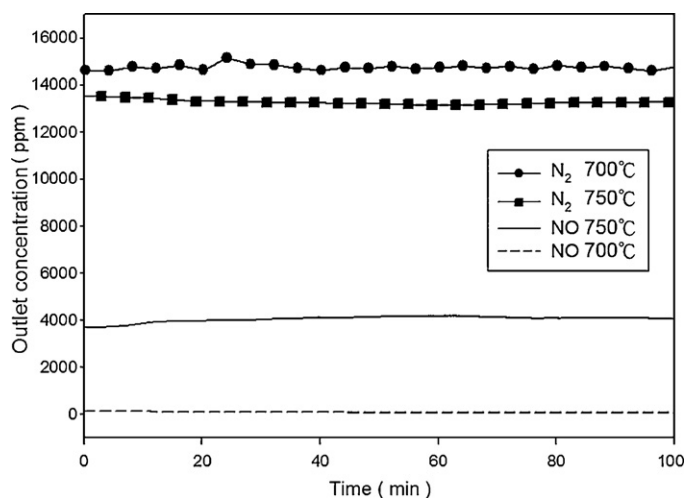
The above results show that all exhaust components of O<sub>2</sub>, H<sub>2</sub>O and CO<sub>2</sub> have beneficial effect on NO conversion. Especially, increasing O<sub>2</sub> content greatly increases NO conversion, noting that increasing O<sub>2</sub> content from 2 to 10% in the simulated exhaust increases NO conversion from 18.0 to 69.4% as shown in Fig. 3. These are beneficial for the application of SOFCs to NO removal from lean-burn engine exhausts under practical conditions. Notably, LSM has been shown to be capable of functioning properly after sulfur treatment as a catalyst for lean NO<sub>x</sub> trap [25]—that is, LSM has good sulfur resistance.

#### 3.4. Effect of generated voltage on NO conversion

Fig. 5 shows that the NO conversion decreases slightly with a decrease of the generated voltage from the OCV at 1.1 V to the fixed voltage at 0.4 V, noting that the generated current density increases from zero at OCV to 245.1 mA cm<sup>-2</sup> at the fixed voltage of 0.4 V. This



**Fig. 5.** Effect of generated voltage on NO conversion ( $\Delta$ NO) during steady state in constant-voltage test at 800 °C with cathode gases of 10% O<sub>2</sub>, 10% H<sub>2</sub>O, 10% CO<sub>2</sub> and 2000 ppm NO. OCV: open circuit voltage.



**Fig. 6.** Concentration profiles of outlet gases with 3% NO inlet during steady state in constant-voltage test at OCV.

indicates that the variation of the generated voltage has only very small effect on NO conversion; notably, the effect of the generated current density on NO conversion is especially small. This is beneficial for keeping a constant NO treating efficiency since a variation of the exhaust composition can vary the generated voltage or the generated current density.

#### 3.5. Effect of temperature on NO conversion

Fig. 6 shows that, with 3% NO inlet and with the SOFCs operating at the OCV, only N<sub>2</sub> was detected in the gas product during the constant-voltage tests at 700 and 750 °C. Table 3 shows that NO is almost completely converted at 700 °C. Notably, NO conversion decreases only slightly when the generated voltage is smaller than that at OCV, a behavior similar to that shown in Fig. 5. The use of OCV for NO conversion is to demonstrate that NO decomposition under OCV can be very effective to convert NO to N<sub>2</sub>. The extent of NO decomposition under a fixed voltage smaller than OCV is smaller than that at OCV according to NO conversion. This indicates that the voltage may be the main driving force for NO decomposition. However, the practical usage of OCV for NO conversion needs further studies for clarification.

Additionally, the amount of the converted NO is two times that of the formed N<sub>2</sub>, within the uncertainty range of the measurements. This indicates that NO reaction is NO decomposition with N<sub>2</sub> formation:



Therefore, NO was completely converted to N<sub>2</sub> without the formation of other nitrogen-containing species. Notably, N<sub>2</sub>O is not stable above 500 °C and quantitatively decomposed in the gas phase (without catalyst) at the high temperature of this study; in fact, no N<sub>2</sub>O could be detected during the GC measurements in this work.

Table 3 also shows that the NO conversion increases as the temperature decreases from 750 to 700 °C. Although the voltage also increases with decreasing temperature and an increased voltage

**Table 3**  
Effect of temperature on NO conversion and N<sub>2</sub> formation with 3% NO fed to the cathode under OCV condition.

Temperature (°C)	OCV (V)	NO conversion (%)	NO converted (ppm)	N <sub>2</sub> formed (ppm)
700	0.65	99.8	29,940	14,900
750	0.50	87.6	26,290	13,100

can increase the NO conversion, the effect of voltage is relatively small as indicated above. Thus, the observed increase of the NO conversion should be mostly due to a temperature effect. This enhanced NO conversion at lower temperature is beneficial for NO removal using the SOFCs since an SOFC can be heated to a lower operating temperature faster and with less heating requirement. Notably, lower SOFC operating temperature should also be beneficial for the stability due to thermal cycling.

#### 4. Conclusions

The results in this work demonstrate that, when the O<sub>2</sub> content in the simulated exhaust increases from 2 to 10%, the NO conversion increases from 18% to about 70%. An increase of NO concentration from 600 to 5000 ppm also increases the NO conversion considerably. Adding 10% H<sub>2</sub>O, increasing CO<sub>2</sub> content from 0 to 20% and increasing the generated voltage from the fixed voltage at 0.4 V to the OCV at 1.1 V all increase the NO conversion, although only slightly. All the exhaust components of O<sub>2</sub>, NO, H<sub>2</sub>O and CO<sub>2</sub> have beneficial effect on the NO conversion. It is also confirmed that NO can be completely converted to N<sub>2</sub> without the formation of other nitrogen-containing species.

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