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Reversible Solid Oxide Cells: Selection of Fuel Electrode Materials for Improved Performance and Durability

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Different fuel electrodes for reversible solid oxide cells (r-SOC) are investigated with the aim of improving performance in both solid oxide electrolysis cell (SOEC) and solid oxide fuel cell (SOFC) modes, and durability in reversible operation mode. Electrodes based on gadolinium-doped ceria (GDC) as a mixed ionic electronic conductor, and lanthanum-doped strontium titanate (LST) as an electronic conductor are selected. The current-voltage characteristics of r-SOC single cells, and their cycling durability up to 1000 cycles are evaluated. LST-GDC co-impregnated with Ni and GDC prove to be highly durable in reversible operation, as a suitable fuel electrode material for r-SOCs.

Introduction

Reversible solid oxide cells (r-SOCs) are attractive electrochemical energy devices that can operate as both SOFCs for power generation, and SOECs for steam electrolysis (1). Renewable power generation, such as wind and solar, depends on daily weather conditions, so that it is desirable to adjust the supply of fluctuating electricity to match demand using reversible energy devices such as r-SOCs. The operational concept of r-SOCs is schematically described in Fig. 1. Since similar materials can be used for fuel electrodes in SOECs and SOFCs, such materials may also be applied as electrode materials in r-SOCs. The electrolyte can be a dense material of yttria-stabilized zirconia (YSZ) or scandia-stabilized zirconia (ScSZ), and the fuel electrode material could be, e.g., a porous cermet material of Ni and YSZ or ScSZ (Ni-YSZ or Ni-ScSZ). The perovskite-type oxides such as lanthanum strontium cobalt ferrite and lanthanum strontium manganite (LSM) and are commonly used as air electrode materials.

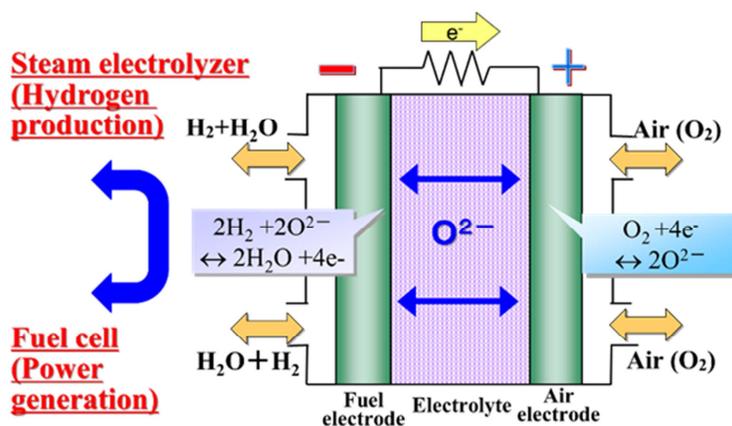


Figure 1. Concept of the reversible solid oxide cell (r-SOC).

In both operation modes, humidified hydrogen at relatively high water vapor partial pressure may be supplied to the fuel electrodes, such that more durable materials, stable in water-vapor-rich atmosphere, should be used in r-SOCs. It is therefore desirable to use a redox-tolerant material for r-SOC fuel electrodes. Among various electron-conducting oxides, SrTiO₃, a perovskite oxide, is redox stable under SOFC operation conditions, and (La_xSr_{1-x})TiO₃ (LST) doped with La acting as a donor at the A site of the perovskite crystal structure is reported to exhibit high electronic conductivity (2,3). In addition, highly dispersed GDC, a mixed ionic electronic conductor under fuel electrode conditions, is expected to expand the electrochemical reaction area, and to suppress carbon deposition (4-6). The impregnation procedure is also applied to improve the electrochemical characteristics of SOFCs by highly dispersing catalyst particles on a stable backbone structure to provide durability against redox cycling. By using a mixture of LST (La_{0.1}Sr_{0.9}TiO₃) and GDC (Gd_{0.1}Ce_{0.9}O₂) as the fuel electrode backbone and by applying the co-impregnation procedure of Ni and GDC, fuel electrodes with highly dispersed nanoparticles of metallic Ni catalysts and GDC, denoted as Ni-GDC/LST-GDC, can be fabricated (7-9).

The aim of this study is to examine the electrochemical properties and durability of r-SOCs using alternative fuel electrode materials. Durability tests are conducted using both the individual constituent materials, and the composite materials as the fuel electrodes in order to identify technical issues, namely LST, GDC, LST-GDC, co-impregnated Ni-GDC/LST-GDC, besides Ni-ScSZ.

Experimental

Various r-SOCs were prepared using conventional and alternative fuel electrode materials. Ni-cermet with ScSZ was used for the fuel electrode as the conventional material reference; dense 200 μm thick ScSZ membrane for the electrolyte; and La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O₃ (LSCF) air electrode with GDC (Gd_{0.1}Ce_{0.9}O₂) acting as a buffer layer between the ScSZ electrolyte and the LSCF air electrode. As alternative fuel electrode materials, LST (La_{0.1}Sr_{0.9}TiO₃) and GDC were used for the fuel electrode backbone. By applying the co-impregnation procedure of Ni and GDC, fuel electrodes with highly dispersed nanoparticles of Ni catalysts and GDC, denoted as

Ni-GDC/LST-GDC, were prepared. A Pt-based reference electrode was deposited beside the air electrode, such that the fuel electrode voltage (potential) was measured as the voltage between the fuel and reference electrodes.

A schematic of the experimental setup for electrochemical measurements is shown in Fig. 2. After a cell was set in the evaluation system, the cell in the ceramic tube furnace was heated to 1000 °C, over 5 h, to melt the Pyrex glass rings between the alumina rings and the alumina tubes to form gas seals (10). The fuel electrode was then subjected to a reduction process (from NiO to Ni) in 3%-humidified hydrogen gas for 1 h. After the reduction process, the cell temperature was lowered to 800 °C.

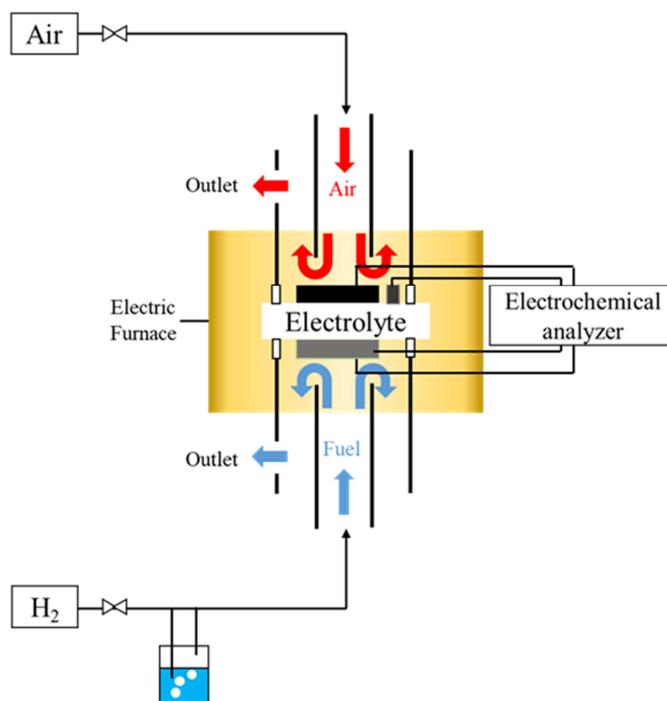


Figure 2. A schematic of the experimental setup for electrochemical measurements.

Current-voltage characteristics were measured at 800 °C. Air was supplied to the air electrode, while 50%-humidified hydrogen gas was supplied to the fuel electrode. The current density in the SOFC and SOEC modes is denoted as positive and negative values, respectively.

To evaluate the durability of the fuel electrodes against repeated reversal of the operation mode, r-SOC cycling durability tests were conducted. As shown in Fig. 3, the current density was repeatedly switched between $\pm 0.2 \text{ A cm}^{-2}$, up to 1000 cycles at 800 °C.

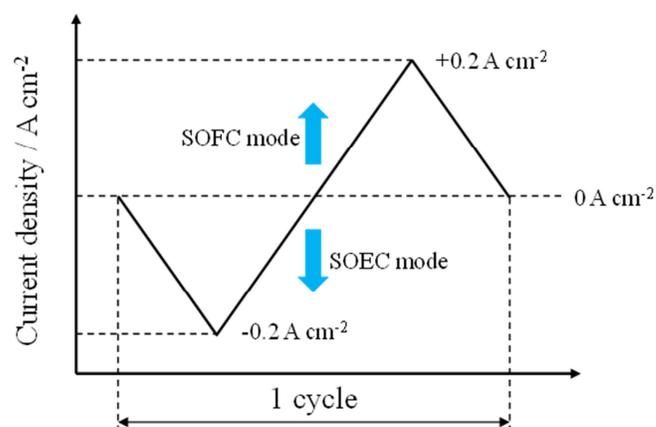


Figure 3. A schematic of one r-SOC cycle by varying current density.

Results and Discussion

Figure 4 shows fuel electrode voltage (potential) as a function of current density in both SOFC and SOEC modes. Absolute values of fuel electrode voltage are shown, measured against the reference electrode in air on the air electrode side. The cells with the Ni-ScSZ cermet, GDC, LST, LST-GDC, and Ni-GDC/LST-GDC could be operated in both modes. In the SOFC mode, the conventional Ni-zirconia cermet exhibited the highest fuel electrode voltage among these materials studied. In contrast, in the SOEC mode, the GDC-containing materials exhibited lower (i.e. better) fuel electrode voltage compared to Ni-zirconia cermet even without Ni catalysts, indicating enhanced electrocatalytic activity of mixed-conducting GDC for the electrode reactions with water vapor at the fuel electrodes.

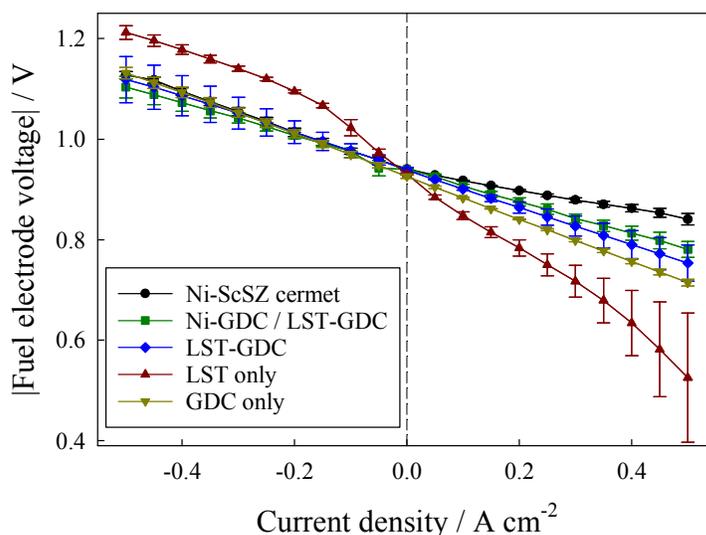


Figure 4. Fuel electrode voltage (absolute value) as a function of current density of r-SOCs using various fuel electrode materials, measured at 800 °C.

The r-SOC cycling durability tests were conducted by switching between the SOEC and SOFC modes for three representative types of fuel electrodes: the conventional Ni-ScSZ cermet; the LST-GDC (without Ni); and the Ni-GDC/LST-GDC. The fuel electrode voltage with respect to the reference electrode is shown in Fig. 5. The data is plotted after every 50 cycles. The data above 1.0 V corresponds to the fuel electrode voltage in the SOEC mode (at -0.2 A cm^{-2}), and the data below 1.0 V to the fuel electrode voltage in the SOFC mode (at $+0.2 \text{ A cm}^{-2}$). The fuel electrode voltage of the Ni-ScSZ cermet increased in the SOEC mode, and decreased in the SOFC mode over the course of the durability test, corresponding to substantial degradation of the fuel electrode performance even though further improvement may be possible. The Ni-GDC/LST-GDC fuel electrode exhibited much higher durability than the Ni-ScSZ cermet fuel electrode, although the overvoltages slightly changed in both SOEC and SOFC modes. These results indicate that the co-impregnated Ni-GDC/LST-GDC is a promising fuel electrode material for r-SOCs. The porous LST-GDC backbone even without Ni catalysts is also catalytically active with sufficient reversible cycling durability.

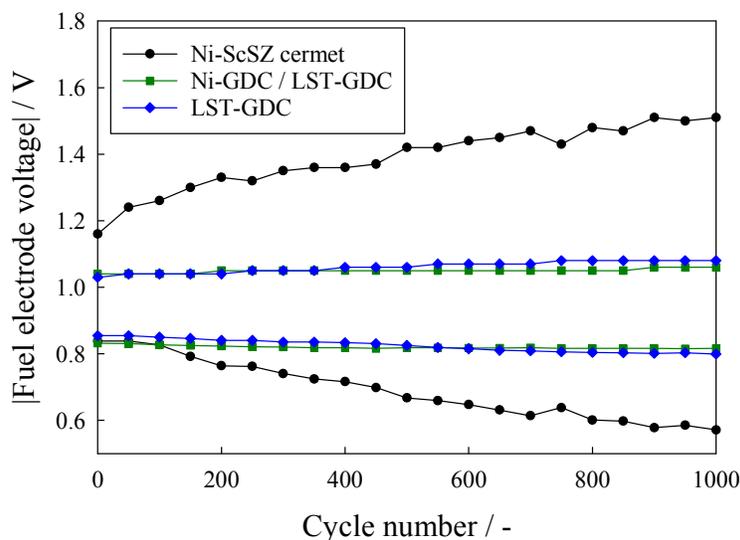


Figure 5. Fuel electrode voltage (absolute value) up to 1000 cycles of the Ni-ScSZ cermet fuel electrode, the Ni-GDC/LST-GDC fuel electrode, and the LST-GDC fuel electrode, at 800 °C.

Conclusions

A series of r-SOCs was prepared with GDC, LST, LST-GDC, and Ni-GDC/LST-GDC fuel electrodes. Compared with the conventional Ni-zirconia cermet fuel electrodes, the LST-GDC based fuel electrodes co-impregnated with Ni and GDC were found to have improved I-V characteristics, and better r-SOC cycling durability over 1000 cycles. This may be partly attributed to the high catalytic activity of GDC. The LST-GDC based electrodes prove to be highly durable in cyclic operations. As such, LST-GDC

co-impregnated with Ni and GDC is shown to be a suitable fuel electrode material for r-SOCs. Further detailed analysis on long-term durability and microstructural stability is in progress.

Acknowledgment

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