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# Thermal cycling of anode supported solid oxide fuel cells under various conditions: Electrical anode protection

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## ABSTRACT

Anode supported cells were thermal cycled in the temperature range 200–750 °C under various conditions: (1) without hydrogen supply, (2) with hydrogen supply, and (3) applying negative current (without hydrogen). Power density and impedance spectra were measured after several cycles and cycling durability was compared for each condition. Post-material analyses were conducted on the anode layers of the thermal cycled cells, using scanning electron microscopy, energy dispersive spectroscopy, and Brunauer–Emmett–Teller. There were no appreciable changes in performance and anode microstructure after the cycling test in the case of ‘Condition 2 and 3’; however, the cell was significantly degraded after a few cycles under ‘Condition 1 (the slow rate)’, due to re-oxidation of Ni anode. The results indicate that the electrical method (applying a negative current) has an effect for anode protection equivalent to that of the conventional chemical method (cover gas supply). This electrical method is preferred, particularly in consideration of emergency shut-down events with fuel loss and fuel economy penalty.

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## Introduction

Solid oxide fuel cells (SOFCs) typically operate above ~700 °C, and this high operating temperature provides several advantages such as high efficiency and fuel flexibility [1]. This type of fuel cell system is appropriate for a stationary electric power application rather than portable one; so long term stability under high temperature operation is a requirement

for its commercialization [2,3]. SOFC durability has been extensively studied from small single cells to short stack levels, with focus on thermal, load and redox cycling. This is because cycling performance must be good to achieve practical application and commercialization [4–18]. Fuel cell systems must be able to cool down and restart readily (i.e., facile thermal-cycling). This is particularly important when power is not required or in the event of emergency shutdown. Due to its high-temperature operation, a SOFC stack undergoes a

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long procedure of thermal cycling over a wide temperature range. As thermal cycling occurs, contact resistance between electrode and electrolyte, and/or cell and interconnector, may significantly increase because of mismatched thermal expansion coefficients, which eventually results in thermal stress [19–22]. Most research about thermal cycling of SOFCs has focused on thermal stress issues: the cycling tests were usually conducted with a supply of cover gas or hydrogen fuel.

The conventional anode material of SOFCs (Ni + YSZ cermet) should be protected from Ni re-oxidation (which may result in anode volume expansion and cracks) during thermal cycling to prevent anode performance degradation. Conventionally, a cover gas (usually hydrogen diluted with nitrogen) is supplied to the anode during heating and cooling, to maintain the equilibrium potentials. If the sealant is not perfect, or if pin holes exist in the electrolyte, the cell voltage can drop below the equilibrium potential unless cover gas is continuously supplied during thermal cycling. The equilibrium potential (Ni/NiO) can be calculated using Eq. (1) [23]:

$$OCV = \frac{1}{2F} \left( \frac{R \ln \left( \frac{p_{O_2}^{cathode}}{2} \right)}{2} T - \Delta G_{Ni/NiO}^{\circ}(T) \right) \quad (1)$$

where T is temperature in Kelvin, R the gas constant, F the Faraday constant, and  $p_{O_2}^{cathode}$  the partial pressure of oxygen at the cathode side. Fig. 1 shows the calculated Nernst potential (open circuit voltage) as a function of temperature for the Ni/NiO equilibrium in the case that the partial pressure of oxygen at the cathode is ~0.21 atm. Because the Tammann temperature (at which oxygen mobility and reactivity become appreciable) of nickel is 590 °C, it is not necessary to maintain the equilibrium potential in the low temperature range (<550 °C) due to the slow kinetics of Ni re-oxidation [24].

The conventional anode protection method using cover gases is not efficient in regard to maintenance costs (fuel economy penalty) and adds system complexity. Moreover, in emergency shut-down or fuel-loss events, the Ni anode can be re-oxidized because of failure to maintain the Ni/NiO equilibrium potential. In addition to this chemical method (cover gas supply), an electrical method can also be utilized to maintain the equilibrium potential for Ni, simply by using a DC power supply. In this case, the cell voltage is electrically maintained with the help of a DC power supply. Oxygen and/

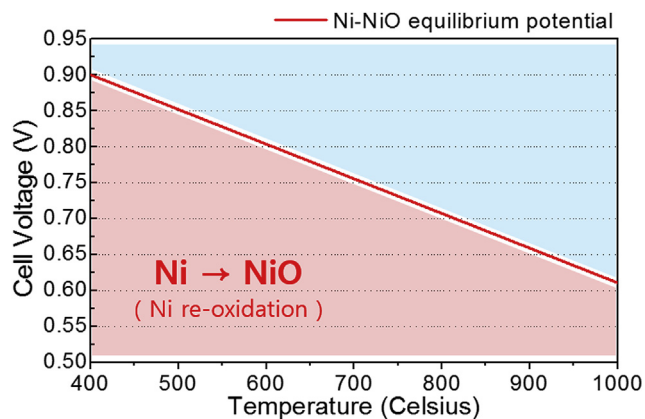


Fig. 1 – Nernst potential vs. temperature for the Ni/NiO equilibrium potential.

or water vapor molecules on the fuel electrode (Ni electrode) are electrochemically transported to the air electrode via oxygen-ion conducting electrolyte such as YSZ (yttria-stabilized zirconia), to maintain the potential above equilibrium. Fig. 2(a) and (b) compare electrons/oxygen ions transport during fuel cell operation and electrical protection (applying a negative current). The basic concept of this electrical method was proposed in the patents [25,26].

In this study, we investigated SOFC thermal cycling durability under various conditions. Three different conditions were set: (1) without supply of hydrogen (cover gas), (2) with supply of hydrogen gas and (3) applying a negative current using potentiostat without hydrogen (cover gas). This allows the cell voltage to be in the safe range by electrochemically transporting oxygen from the anode to the cathode through the electrolyte. These three conditions were separately applied to three identical anode supported cells. It is noted that button size cells were prepared because the purpose of this research is to investigate the validity of the electrical protection on SOFC durability during thermal cycling, in comparison with the chemical protection. If larger area cells are tested with unit-cell-assembly components (such as ceramic-glass-type sealant and metallic interconnectors), we should consider additional degradation factors that originate from thermal stress, metallic part corrosion, and gas cross-over through broken sealant, as thermal cycling proceeds. Power density and impedance spectra were measured and compared before and after cycling under three different conditions, to trace any increase in ohmic and non-ohmic resistance. Post-material analyses were also conducted on the thermally cycled cells, with focus on the anode parts, using scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and Brunauer–Emmett–Teller (BET). To our

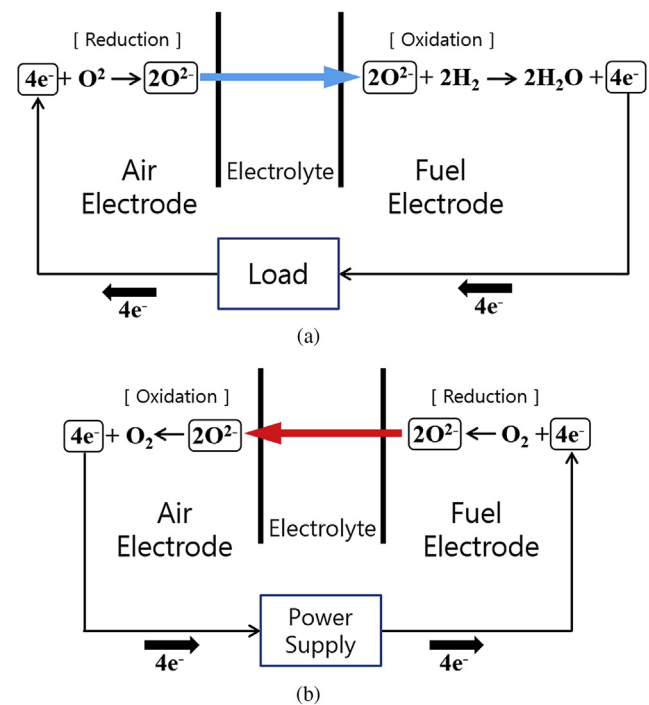


Fig. 2 – Schematic of electron and oxygen ions transport during fuel cell operation (a) and electrical protection (b).

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