<table>
<thead>
<tr>
<th><strong>Title</strong></th>
<th>A promising Ni-Fe bimetallic anode for intermediate-temperature SOFC based on Gd-doped ceria electrolyte</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Author(s)</strong></td>
<td>Fu, C. F.; Chan, S. H.; Ge, Xiaoming; Liu, Q. L.; Pasciak, G.</td>
</tr>
<tr>
<td><strong>Date</strong></td>
<td>2011</td>
</tr>
<tr>
<td><strong>URL</strong></td>
<td><a href="http://hdl.handle.net/10220/8141">http://hdl.handle.net/10220/8141</a></td>
</tr>
<tr>
<td><strong>Rights</strong></td>
<td>© 2011 Hydrogen Energy Publications, LLC. This is the author created version of a work that has been peer reviewed and accepted for publication in International Journal of Hydrogen Energy, published by Elsevier on behalf of Hydrogen Energy Publications, LLC. It incorporates referee's comments but changes resulting from the publishing process, such as copyediting, structural formatting, may not be reflected in this document. The published version is available at: [DOI: <a href="http://dx.doi.org/10.1016/j.ijhydene.2011.07.119">http://dx.doi.org/10.1016/j.ijhydene.2011.07.119</a> ].</td>
</tr>
</tbody>
</table>
A promising Ni-Fe bimetallic anode for intermediate-temperature SOFC

based on Gd-doped ceria electrolyte

C.J. Fu* a, S.H. Chan b, X.M. Ge b, Q.L. Liu b and G. Pasciak c

aSchool of Materials Science & Engineering, Helongjiang Institute of Science & Technology, 1 Tangchang Road, Harbin 150027, China

bSchool of Mechanical and Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

cElectrotechnical Institute, Division of Electrotechnology and Materials Science, M. Sklodowskiej Curie 55/61, 50-369 Wroclaw, Poland

Abstract

Anode-supported solid oxide fuel cells (SOFC) based on Ni-Fe bimetal and gadolinia-doped ceria (GDC) composite anode were fabricated and evaluated in the intermediate- and low-temperature range. Ni0.75Fe0.25-GDC anode substrate and GDC electrolyte bilayer were prepared by the multi-layered aqueous tape casting method. The single cell performance was characterized with La0.6Sr0.4Co0.2Fe0.8O3-GDC (LSCF-GDC) composite cathode. The maximum power density reached 330, 567, 835 and 1333 mW cm² at 500, 550, 600 and 650°C, respectively. Good long-term performance stability has been achieved at 600°C for up to 100 h. The improved single cell performance was achieved in the reduced temperature after the long-term stability test. The maximum power density registered 185 and 293 mW cm² at 400 and 450°C, respectively. The impedance spectra fitting results of the test cell revealed that the improved cell performance was attributed to the much lower electrochemical reaction resistance. XRD and SEM examination indicated that the outstanding performance of the single cell seemed to arise from the

* Corresponding author
E-mail address: fcj_hit@163.com (CJ Fu)
optimized composition and excellent microstructure of \( \text{Ni}_{0.75}\text{Fe}_{0.25} \)-GDC anode, as well as the improved stability of the anode microstructure with prolonged testing time.

Keywords: IT-SOFC; Anode; Ni-Fe bimetal; Electrolyte; GDC

1. Introduction

Solid oxide fuel cell (SOFC) is a promising clean energy technology for the short-to-medium-term substitution of fossil fuels [1, 2]. In recent years, the focus of SOFC development has been focused on lowering the operating temperature of SOFCs to 500–800°C [3]. Motivation for lowering the operation temperature of SOFC is primarily driven by the cost and durability, because by lowering the temperature of operation, the problems associated with thermal cycling and performance degradation caused by the interdiffusion or reaction of the individual components, can be alleviated, thus enhancing the durability of SOFC systems. Operation at lower temperature, however, creates a number of problems that are associated with the increase in the electrolyte resistance and decrease in the rate of electrocatalytic reactions [4]. Decreasing the operating temperature of SOFC increases the electrode polarization, thus becoming performance-limiting factor as the apparent activation energies for the interfacial processes are known to be higher than the ionic transport in solid state electrolytes [5, 6]. Therefore, it is necessary to continuously search for novel cathode and anode materials having superior electrocatalytic activity in the intermediate temperature range [7, 8]. Previously, cathodes received much more attention because the polarization loss of the cathode is considered to be the major issue affecting SOFC performance at the low operating temperatures [9]. Effects of the anode on SOFC performance in the reduced temperature were always neglected.

Goodenough et al. [10, 11] has discussed the requirements for a suitable anode material, namely, the compound must have adequate electronic conductivity implying that in the reducing atmosphere at the anode, catalytic activity requires that the mixed valence redox couple can accept electrons from \( \text{H}_2 \) or the hydrocarbon fuel in order to induce dissociation on the oxide surface whose ionic conductivity is
determined by oxygen ions diffuse rapidly from the electrolyte to the surface of the anode. Currently, the composition of Ni with an oxide-ion conductor such as yttria-stablized zirconia (YSZ) has been widely used as SOFC anode, since it has good electrical conductivity and high catalytic ability towards $\text{H}_2$ oxidation [12]. However, structural and long-term performance stability of Ni-YSZ cermet anodes is critical issues for the development of SOFC anode [13]. The most predominant microstructure changes of Ni-YSZ anode is the agglomeration and coarsening of Ni phase [14]. Since the electrocatalytic activity of the anode can have a significant effect on the electrochemical performance of SOFC, development of high performance anodes beyond pure Ni are desirable to lower the overall anode polarization resistance and increase the cell power density, thus reducing the cost of the commercial SOFC stack [15].

Recently, with the development of metallic supported SOFC, increasingly more attention has been paid to Ni-Fe bimetal anodes. Ishihara and co-workers [16, 17] reported the development of LSGM-based cells with Ni-Fe alloying anode to improve the anode catalytic activity. They fabricated a few micrometers thick LSGM/SDC bilayer electrolyte on the dense NiO-Fe$_2$O$_3$ metallic support by ablation pulsed laser deposition (PLD) method and using $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ (SSC) as cathode. The single cell demonstrated a quick start-up within 5 min and generated the highest power density of 0.95 W cm$^{-2}$ at 600°C and 0.1 W cm$^{-2}$ at 400°C. While using NiO-Fe$_3$O$_4$ (Ni-Fe, 90:10 in molar ratio) and Sm-doped ceria (SDC) composite anode, the maximum power density of the single cell achieved 1.95 W cm$^{-2}$ at 600°C, 0.612 W cm$^{-2}$ at 500°C and 0.08 W cm$^{-2}$ at 400°C [18]. Fiuza et al. [19] studied the electrocatalytic activity of the anode composite Ni-Fe supported on YSZ running on ethanol fuel. It presented a good compromise between satisfactory catalytic activity and carbon formation with Ni as the reforming and oxidation catalyst, and Fe as the conducting and resistant phase for carbon formation. Lu et al. [15] optimized the Ni-Fe composition and reported the performance of Ni-Fe alloy anode on YSZ electrolyte support (200μm in thickness). The single cell with YSZ-Ni$_{0.8}$Fe$_{0.2}$ anode and YSZ-La$_{0.8}$Sr$_{0.2}$MnO$_3$ cathode showed an impressive performance with a maximum power density of 1.43W cm$^{-2}$ at 800°C and 0.62 Wcm$^{-2}$ at 700°C [20]. Park and Virkar [21] studied the performance of the cell with Ni-Fe (70:30 in molar ratio)
and Gd_{0.1}\text{Ce}_{0.9}O_{2-\delta} (GDC) composites as SOFC anode substrate, GDC as electrolyte and Sr-doped LaCoO$_3$ as cathode. With H$_2$ as fuel, the system showed the maximum power density of 0.68 Wcm$^{-2}$ at 650°C. Until now, only good electrocatalytic activity of Ni-Fe bimetallic anode in the intermediate- and low-temperature range has been reported. However, the reduction characteristics of Ni-Fe bimetallic anode in the fuel environment, especially with water, should be paid more attention because, on one hand, water is the anode reaction products; on the other hand, it is also the component of the anode fuel gas in order to reduce the interfacial polarization resistance of hydrogen adsorption. In addition, the long-term performance stability of Ni-Fe bimetallic anode under SOFC operation conditions has yet to be verified, which can be an alternate to Ni anode.

This study focused on the improvement of intermediate-temperature SOFC performance by controlling the microstructure of Ni$_{0.75}$Fe$_{0.25}$-GDC anode with the conventional ceramic fabrication techniques. The anode substrate was prepared based on GDC electrolyte by cost-effective aqueous-based tape casting method. The reduction characteristics of the Ni-Fe anode were characterized under SOFC operating conditions. The electrochemical performance and long-term stability of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC were evaluated as well.

2. **Experimental**

2.1 **Multilayered aqueous-based tape-casting anode/ electrolyte bilayer**

The starting ceramic material used for the fabrication of green tapes was a powder mixture consisting of Gd$_{0.1}\text{Ce}_{0.9}O_{1.95}$ (GDC, Nextech, US), Fe$_2$O$_3$ and NiO (J.T. Baker, US). The anode powders were mixed with 35 wt.% GDC and 65 wt.% NiO-Fe$_2$O$_3$ (75 at.% NiO and 12.5 at.% Fe$_2$O$_3$) by ball milling in isopropanol for 4 h. 5wt.% graphite was also added to adjust the thermal shrinkage of the anode substrate, as well as to facilitate the gas permeation in anode. The as-prepared slip was dried overnight until the solvent was totally evaporated. Details of the sample preparation and optimized slip formula for aqueous-based tape casting Ni$_{0.75}$Fe$_{0.25}$-GDC anode substrate and GDC electrolyte were presented in our work [22].
De-ionized water was used as the solvent. Polyvinyl alcohol (88% partially hydrolyzed) and Polyethylene glycol (PEG400) were selected as the binder and plasticizer, respectively. 2,4,7,9-tetramethyl-5-decylene-4,7-diol ethoxylate, was added to adjust the wetting properties of the suspension. Octanol was used as the deformer and glycerol as the anti-adhesive. Dispersant acrylic acid, ammonium salt polymer was chosen as a dispersing agent with the pH value controlled at around 9~10. The anode materials were mixed and ball-milled with solvent and dispersant for 24 h and ball-milled again with a binder solution for another 24 h to yield the proper slip rheology. Tape casting was performed using a lab tape casting machine (Richard E Mistler, Inc., TTC-1200). The thickness of the green tapes of the electrolyte/anode bilayer was ~1 mm after drying.

2.2 Single cell fabrication

The as-prepared green tapes were shaped and co-sintered at 1400ºC for 5 h to obtain Ni$_{0.75}$Fe$_{0.25}$-GDC anode/GDC electrolyte bi-layer. The gradient composite cathode with the optimized composition was applied on the surface of electrolyte by screen printing method [23]. The cathode functional layer was consist of La$_{0.6}$Sr$_{0.4}$Co$_{0.2}$Fe$_{0.8}$O$_3$ (LSCF, Fuel Cell Materials, US) and GDC in 60:40 by weight. After drying of the printed cathode functional layer in 80ºC oven, LSCF paste was printed on the top of it to form the cathode current collector layer and then drying again. Thus-prepared cathode was finally sintered at 900ºC for 2 h to complete the fabrication of SOFC single cells.

2.3 Single cell characterization

The single cell testing was conducted in the temperature range of 400 to 650ºC in humidified H$_2$ with 3 vol.% H$_2$O and air. The effective area of the cathode was ~0.5 cm$^2$. The flow rate of the fuel and air was set at 50 and 100 sccm, respectively. Electrochemical measurements were conducted using the Solartron Electrochemical Interface (1255B) in conjunction with Solartron Frequency Response Analyzer (1287). The polarization curves were obtained by using a potentiostat with a scan rate of 1 mV s$^{-1}$. The electrochemical impedance spectra were measured under open circuit conditions in the frequency range of
0.1 Hz to 1 MHz. The impedance was analyzed in terms of equivalent circuit by using ZSimWin software. Long-term performance stability of the anode-supported SOFC was examined under the constant potential of 0.16 V vs. open circuit voltage (OCV) up to 100 h. The microstructure of the tested single cell was characterized using a scanning electron microscope (SEM, Jeol 5600LV, Japan). X-ray diffraction (XRD, Philips MPD 1880) measurements using Cu Kα1 radiation (=1.54060Å) have been performed to examine the reduced anode composition at room temperature.

3. Results and discussions

3.1 Microstructure of the tested single cell

SEM images of the tested single cell after reduction and long-term stability test are given in Fig. 1. Fig. 1a shows the multi-layer cell structure. GDC electrolyte is sandwiched by a porous LSCF-GDC composite cathode (bottom layer) and a porous Ni₀.₇₅Fe₀.₂₅-GDC anode (top layer). The thickness of the electrolyte and cathode is ~14 μm and ~70 μm, respectively. The electrolyte seems to be homogenous in thickness. A crack-free dense GDC electrolyte film has been successfully deposited on the Ni₀.₇₅Fe₀.₂₅-GDC anode substrate by a co-sintering technique without open pores seen in Fig. 1b. There are a very few discrete pinholes distributed across grain boundaries with a diameter of less than 0.5 μm. Good adhesion was achieved between the electrolyte and the electrodes. The inset of Fig. 1b is the GDC electrolyte surface microstructure. It demonstrates that GDC electrolyte film prepared by multi-layered tape casting method could be densely co-sintered at 1400°C. SEM image of the anode after reduction is shown in Fig. 1c. Ni₀.₇₅Fe₀.₂₅-GDC anode presents a very different microstructure from the usual Ni-GDC anode [22]. The anode porosity appears to be in bimodal distribution. Coarse pores are in the order of 10 μm caused by the pore-former burnt up. The reduction of nickel and iron oxides resulted in the formation of very fine pores. This special kind of porous microstructure of Ni₀.₇₅Fe₀.₂₅-GDC anode not only allows easy transport of fuel, but also provides much larger surface area for anode chemical reactions to occur, which greatly enhanced the single cell performance. Compared with the anode, the cathodes presents a more uniformly
distributed porous microstructure, as shown in Fig. 1d. According to the SEM images, after the long-term performance test, the single cell exhibited a good microstructure stability and homogeneity. The higher cell performance of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC, to a large degree, might be due to the special bimodal pore distribution microstructure of the anode obtained by the optimized cell components and fabrication method.

### 3.2 Anode composition analysis

Figure 2 shows the XRD patterns of Ni$_{0.75}$Fe$_{0.25}$-GDC anode after reduction. The highest peaks in this trace correspond to a face-centered cubic (FCC) structure and are consistent with the presence of Pt (JCPDS 004-0802), which was induced because Pt paste was used as the current collector of anode. The resulting anode was composed of CeO$_2$ (Fluorite structure, JCPDS 043-1002) and the solid solution of Ni and Fe as the major phases. All the diffraction peaks of CeO$_2$ in the anode shifted to the low-angle side indicating that gadolinium doping resulted in an expansion of the unit cell of CeO$_2$ and the direct formation of solid solution. However, it cannot be determined whether all the dopants are in the form of the solid solution or partially present as phases undetectable with XRD [24]. Iron and nickel are adjacent to each other in the periodic table, and easily form widely solid solutions and intermetallic phases. Solid solutions of Awaruite FeNi$_3$ (JCPDS 38-0419) and Kamacite FeNi (JCPDS 18-0645) were both found in FCC structure. The appearance of FeNi phase maybe due to the volatile of nickel during the process of high temperature co-sintering of electrolyte/anode bilayer resulting in the loss of nickel in the anode. The examination of Fig. 2 also showed small traces of unknown phase which was probably due to the impurity induced during the cell fabrication and test process.

### 3.3 Single cell performance

Figure 3 shows the typical current-voltage curves of the single cell consisting of Ni$_{0.75}$Fe$_{0.25}$-GDC composite anode, GDC electrolyte and LSCF-GDC composite cathode, measured at temperatures between 500 and 650°C. The OCV registered 0.82, 0.87, 0.92 and 0.95 V at 650, 600, 550 and 500°C,
respectively. The maximum power density achieved 330, 567, 835 and 1333 mW cm$^{-2}$ at 500, 550, 600 and 650°C, respectively. The performance of this single cell with Ni$_{0.75}$Fe$_{0.25}$-GDC anode (835 mW cm$^{-2}$ at 600°C) was much higher than those recently reported Ni-GDC anode-supported SOFC. Liu et al. [25] prepared Ni-GDC anode substrate by die-pressing method. The maximum power density of the single cell (Ni-GDC/GDC/LSCF-GDC) was 562 mW cm$^{-2}$ at 600°C. Fu et al. [22] fabricated Ni-GDC anode by the aqueous tape casting method achieved a better performance with the highest power density reached 625 mW cm$^{-2}$ at 600°C with LSCF-GDC composite as cathode. This result further confirmed that iron could greatly enhance the catalytic activity of SOFC anode reaction in the intermediate- and low- temperature range [26].

The long-term performance stability of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC measured at 600°C under SOFC operation conditions is given in Fig. 4. The measurement was conducted for 100 h under the constant potential of -0.71 V (0.16 V vs. OCV). In the early stage of the testing (less than 1.5 h), the current density increased about 6%, from the initial value of 556 to 590 mA cm$^{-2}$. The current density increase could be ascribed to the enlargement of the effective triple phase boundary (TPB) due to the improved contacts between electrodes and GDC grains [27]. In the following 57 h, the current density increased slowly, registered only 16 mA cm$^{-2}$. Then it kept almost unchanged for the last 41 h. These results indicated that Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC not only demonstrated an excellent cell performance, it also maintained an excellent long-term stability in the intermediate-temperature range. It did not appear any of the degradation caused by the microstructural change usually happened in the traditional Ni anode [28].

After the long-term stability test, the single cell performance was further measured in the lower temperature of 400 and 450°C, as shown in Fig. 5. The OCV were 0.98 and 1.02 V at 450 and 400°C, respectively. The maximum power density reached 185 and 293 mW cm$^{-2}$ at 400 and 450°C, respectively. Comparing the power density between 450°C (after long-term stability test) and 500°C (before long-term stability test), the results showed that they were close to each other, which further confirmed activation
effect of electrodes during long-term stability test process. At 400°C, the maximum power density of \( \text{Ni}_{0.75}\text{Fe}_{0.25}\text{-GDC} \) anode supported SOFC was 185 mW cm\(^{-2}\), which is 10% higher than that of \( \text{Ni}\text{-GDC} \) anode supported SOFC at 500°C (168 mW cm\(^{-2}\)) [22]. The higher power density of the \( \text{Ni}_{0.75}\text{Fe}_{0.25}\text{-GDC} \) anode supported SOFC in the low temperature range was likely due to activation of electrodes during the long-term stability test process and iron doping, which greatly improved the low temperature performance of SOFC.

3.4 EIS analysis

The impedance spectra of the single cell (\( \text{Ni}_{0.75}\text{Fe}_{0.25}\text{-GDC/GDC/LSCF-GDC} \)) measured under open circuit conditions between 500 and 650°C are given in Fig. 6a. The best fitting results to the equivalent circuit of \( LR_0(R_1Q_1)(R_2Q_2) \) for all the impedance spectra have been achieved, indicating that at least two different electrode processes corresponding to the high- and low-frequency arcs exist. The high-frequency inductive loop, \( (R_1Q_1) \), is associated with the charge-transfer process on the electrolyte surface near the interface region. The low-frequency inductive loop, \( (R_2Q_2) \), can be attributed to the activation of a passive layer at the electrode surface or adsorption phenomena. The total polarization resistance of the single cell, \( R_p \), is the sum of \( R_1 \) and \( R_2 \).

\[
R_p = R_1 + R_2
\]  

(1)

The inductance of the circuits, \( L \), may be the result of the Pt current/voltage probes or the high frequency phase shift of the electrochemical equipment. The overall ohmic resistance, \( R_\Omega \), includes the electrolyte resistance, the electrodes ohmic resistance, the contact resistance between the electrodes and electrolyte and the leads resistance. The fitted impedance parameters for the single cell are summarized in Table 1. The ohmic resistance \( (R_\Omega) \) of the single cell increased by 2.6 times when the temperature was lowered from 650°C (0.05 \( \Omega \text{ cm}^2 \)) to 500°C (0.18 \( \Omega \text{ cm}^2 \)); while the polarization resistance \( (R_p) \) increased nearly by 10-fold, from 0.068 \( \Omega \text{ cm}^2 \) at 650°C to 0.74 \( \Omega \text{ cm}^2 \) at 500°C. In the \( \text{Ni}_{0.75}\text{Fe}_{0.25}\text{-GDC} \) anode supported
SOFC, the interfacial polarization still contributed significantly to the total cell resistance, though it was in a relatively low value. At 650°C, $R_p$ accounts for nearly 58% of the total cell resistance, which increased to 80% at 500°C. Nevertheless, $R_\Omega$ and $R_p$ of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC were greatly lower than that of Ni-GDC anode supported SOFC, which presented $R_\Omega$ of 0.35 Ω cm$^2$ at 650°C and 0.57 Ω cm$^2$ at 500°C, while $R_p$ was 0.4 Ω cm$^2$ at 650°C and 4.4 Ω cm$^2$ at 500°C, respectively [25]. Thus, it can be concluded that iron doping in SOFC nickel anode could significantly improve the anode catalytic activation and accelerate the reaction process of the electrochemical kinetic at the reduced temperature. The cell impedance at different temperature under open circuit conditions and the corresponding temperature-dependant total cell polarization resistance are given in Fig. 6b. The apparent activation energies of the polarization resistance in the low- and high-frequency range and the total polarization resistance registered 122.5 kJ mol$^{-1}$, 65.8 kJ mol$^{-1}$ and 95.3 kJ mol$^{-1}$, respectively, which are lower than those of Ni-GDC anode supported SOFC [24]. The apparent activation energy of the ohmic resistance was 49.9 kJ mol$^{-1}$. These activation energy results further proved that the reduced single cell performance was due to the sluggish electrochemical kinetics of the electrodes, especially the activation of a passive layer at the electrode surface and adsorption process, in the intermediate-temperature range. Figure 7 shows the EIS comparison of the single cell before and after the long-term performance test at 600°C. $R_\Omega$ and $R_p$ decreased 31% and 25%, respectively, according to Table 1, after 100 h long-term stability test at 600°C, which was consistent with the enhanced performance test results.

EIS of the single cell after long-term stability test at 450 and 400°C are given in Fig. 8. Their EIS fitting parameters are also given in Table 2. $R_p$ of the single cell at 450°C was only 0.84 Ω cm$^2$, which is slightly higher than that of the single cell at 500°C (0.74 Ω cm$^2$); these further confirmed the activation effect of cell polarization. Comparing $R_1$ and $R_2$ values in Table 1 and Table 2, it seems that $R_1$ was always higher than $R_2$ when the test temperature was 550°C or higher, whereas below 550°C, the values of $R_2$ was approximately two-fold higher than those of $R_1$. The lower the test temperature, the larger the difference will be. These fitting results revealed that charge transfer process was retarded when the test temperature
was 550°C or higher, but the electrodes surface adoption process of the reaction gases was very sluggish when the test temperature was lower than 550°C. Therefore, to further reduce the operating temperature of SOFC, it is necessary to develop new electrode materials with even low activation energy for the process of the reaction gases adoption.

**Conclusions**

Ni-Fe bimetallic alloy demonstrated a high activation for the electrochemical oxidation of hydrogen in SOFC intermediate temperature range. The single cell overpotential was effectively suppressed by using Ni$_{0.75}$Fe$_{0.25}$-GDC anode, especially at reduced temperatures. Iron doping not only lowered the single cell ohmic resistance, but also improved the anode microstructure after reduction, which greatly increased the electrochemical reaction areas for hydrogen. A much higher single cell performance was achieved for Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC than that of Ni-GDC anode supported SOFC. At 500°C, $R_{\Omega}$ and $R_p$ of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC was less than 32% and 5%, respectively, compared to the Ni-GDC anode supported SOFC, respectively. Moreover, this kind of bimetallic alloying anode could also keep the anode microstructure stable and efficiently inhibit nickel from coarsening during the long-term performance test. These results indicated that Ni$_{0.75}$Fe$_{0.25}$-GDC was a promising bimetallic anode for intermediate- and low-temperature SOFC.

**Acknowledgements**

This work was supported by Agency for Science, Technology and Research (A*STAR), Singapore and Ministry of Science and Higher Education, Poland.

**References**

**Lists of Figures and Tables**

**Fig.1** Cross-sectional images of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC after reduction and single cell test: (a) multi-layer SOFC structure; (b) electrolyte (inset is the surface microstructure of GDC electrolyte); (c) anode and (d) cathode.
Fig. 2 XRD patterns of Ni$_{0.75}$Fe$_{0.25}$-GDC anodes after reduction and single cell test.

1- Pt
2- GDC
3- FeNi$_3$
4- FeNi
5- Unknown
Fig. 3 Voltages and power density vs. current density plots for Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC from 500°C to 650°C.
Fig. 4 Long-term performance stability of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC at 600°C under the constant potential of 0.16 V vs. OCV.
Fig. 5 Voltages and power density vs. current density plots for Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC at 400°C and 450°C after the long-term stability test of the single cell.
Fig.6 Impedance spectra of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC measured between 500 and 650°C under the open circuit conditions: (a) impedance spectra and (b) Arrhenius plot of the resistances measured at different temperatures.
Fig. 7 Comparison of the impedance spectra for Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC at 600°C before and after the long-term performance test.
Fig. 8 Impedance spectra of Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC at 400 and 450°C under the open circuit conditions after the long-term performance test.
Table 1 – EIS fitting results for Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC between 500 and 650 °C.

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>$R_\Omega$ (Ω cm$^2$)</th>
<th>$Q_1$</th>
<th>$R_1$ (Ω cm$^2$)</th>
<th>$Q_2$</th>
<th>$R_2$ (Ω cm$^2$)</th>
<th>$R_p$ (Ω cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$Y_0$ (S s$^{-1}$ cm$^{-2}$)</td>
<td>$n_1$</td>
<td></td>
<td>$Y_{0.2}$ (S s$^{-1}$ cm$^{-2}$)</td>
<td>$n_2$</td>
<td></td>
</tr>
<tr>
<td>650</td>
<td>0.05</td>
<td>0.57</td>
<td>0.045</td>
<td>0.09</td>
<td>0.023</td>
<td>0.068</td>
</tr>
<tr>
<td>600</td>
<td>0.087</td>
<td>0.19</td>
<td>0.086</td>
<td>0.05</td>
<td>0.049</td>
<td>0.135</td>
</tr>
<tr>
<td>600, 100 h</td>
<td>0.06</td>
<td>0.17</td>
<td>0.064</td>
<td>0.05</td>
<td>0.037</td>
<td>0.101</td>
</tr>
<tr>
<td>550</td>
<td>0.13</td>
<td>0.03</td>
<td>0.202</td>
<td>0.1</td>
<td>0.124</td>
<td>0.326</td>
</tr>
<tr>
<td>500</td>
<td>0.18</td>
<td>0.19</td>
<td>0.221</td>
<td>0.1</td>
<td>0.159</td>
<td>0.74</td>
</tr>
</tbody>
</table>

Table 2 – EIS fitting results for Ni$_{0.75}$Fe$_{0.25}$-GDC anode supported SOFC at 400 and 450 °C after long-term stability test.

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>$R_\Omega$ (Ω cm$^2$)</th>
<th>$Q_1$</th>
<th>$R_1$ (Ω cm$^2$)</th>
<th>$Q_2$</th>
<th>$R_2$ (Ω cm$^2$)</th>
<th>$R_p$ (Ω cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$Y_0$ (S s$^{-1}$ cm$^{-2}$)</td>
<td>$n_1$</td>
<td></td>
<td>$Y_{0.2}$ (S s$^{-1}$ cm$^{-2}$)</td>
<td>$n_2$</td>
<td></td>
</tr>
<tr>
<td>450</td>
<td>0.19</td>
<td>0.07</td>
<td>0.305</td>
<td>0.025</td>
<td>0.78</td>
<td>0.535</td>
</tr>
<tr>
<td>400</td>
<td>0.27</td>
<td>0.13</td>
<td>0.423</td>
<td>0.06</td>
<td>0.69</td>
<td>0.947</td>
</tr>
</tbody>
</table>