Processes Involving in the Temperature Variations in Solid Oxide Fuel Cells In-Situ Analyzed through Electrode-Segmentation Method

Özgür Aydın,a,b Hironori Nakajima,c,∗,z and Tatsumi Kitahara^c

aInternational Research Center for Hydrogen Energy, Kyushu University, Nishi-ku, Fukuoka 819-0395, Japan
bDepartment of Hydrogen Energy Systems, Graduate School of Engineering, Kyushu University, Nishi-ku, Fukuoka 819-0395, Japan
cDepartment of Mechanical Engineering, Faculty of Engineering, Kyushu University, Nishi-ku, Fukuoka 819-0395, Japan

We aim to mitigate the spatial temperature variations contributing to the thermal stresses in solid oxide fuel cells. We thus analyze the involving processes through spatial temperature, current, and impinge variations in-situ measured by the electrode-segmentation method in a microtubular solid oxide. We find that, despite the preheating, the excess air flow commonly supplied in the practical applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.

© The Author(s) 2015. Published by ECS. This is an open access article distributed under the terms of the Creative Commons Attribution 4.0 License (CC BY, http://creativecommons.org/licenses/by/4.0/), which permits unrestricted reuse of the work in any medium, provided the original work is properly cited. [DOI:10.1149/2.0701603jes] All rights reserved.

The thermal stresses are accounted for the mechanical failure of solid oxide fuel cells (SOFC),1–3 To resist the thermal stresses at maximum achievable electrochemical performance, SOFCs are designed in various forms, for instance flat-tubular, tubular, and planar, etc. For in-situ investigating the properties spatially varying regardless of the SOFC form, microtubular SOFCs (mt-SOFCs) are quite practical owing to the simple form. Besides, a mt-SOFC with a small diameter can represent a unit gas flow channel in other forms. We thus elaborate the temperature, current, and concentration variations in mt-SOFCs.

A mt-SOFC fundamentally consists of three main components, anode, electrolyte, and cathode. The cell fabrication requires sequential applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.

The thermal stresses are accounted for the mechanical failure of solid oxide fuel cells (SOFC),1–3 To resist the thermal stresses at maximum achievable electrochemical performance, SOFCs are designed in various forms, for instance flat-tubular, tubular, and planar, etc. For in-situ investigating the properties spatially varying regardless of the SOFC form, microtubular SOFCs (mt-SOFCs) are quite practical owing to the simple form. Besides, a mt-SOFC with a small diameter can represent a unit gas flow channel in other forms. We thus elaborate the temperature, current, and concentration variations in mt-SOFCs.

A mt-SOFC fundamentally consists of three main components, anode, electrolyte, and cathode. The cell fabrication requires sequential applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.

The cell fabrication requires sequential applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.

The cell fabrication requires sequential applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.

The cell fabrication requires sequential applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.

The cell fabrication requires sequential applications for the convective cooling is the prevailing factor on the temperature variations causing a significant temperature gradient in the air inlet region, that poses a high risk of mechanical failure. In terms of the flow configuration, counter-flow shows larger temperature and current variations. The impedance variations clarify the impact of the temperature distribution on the current variations. Namely, high temperature in the fuel upstream accompanied with the high hydrogen concentration boosts the local current density, thus, results in larger Nernst-loss in the downstream wherein temperature is lower as well. We conclude that the excess air flow indirectly contributes to the thermal stresses and thus we recommend the reduction of the excess flow.
The experimental difficulties in measuring the longitudinal temperature and current variations over the cell surface due mainly to the high operation temperature of SOFCs have been leading researchers to create numerical models. Researchers have been developing thermo-electrochemical models by which they have been exploring the longitudinal variations of temperature, current, and concentration of the concerning species. They have been transferring the temperature variations to the Finite Element Method (FEM) models for predicting the induced thermal stresses. Although the thermo-electrochemical models are usually validated with experimental current-voltage (I-V) curves, they are hardly validated in terms of the in-situ measured temperature variations.

For in-situ measuring the longitudinal temperature variations, there have been valuable attempts indeed. Morel et al. have predicted the temperature variations over a planar cell relying on the impact of the thermal stresses. Razbani et al. have reported temperature variations over a planar cell relying on the impact of the thermal stresses. Santarelli et al. measured the local temperature on the ionic conductivity. Morel et al. have predicted the temperature variations. 

In order to alleviate the temperature variations contributing to the thermal stresses, in this study, we explore the effect of the involving processes on the temperature variations in mt-SOFCs. Regarding as the main involving processes, we focus on the current variations and the convective heat transfer due to the fact that air is extensively fed at excess rates for the cooling purpose in the practical systems. Thereby, we analyze the variations with the co- and counter-flow configurations. We conduct the analyses based on the spatial properties in the lateral direction depending on the potential difference between the segments, that might affect the accuracy of the measurements.

In SOFCs, however, the ceramic-based components enable the segmentation on the EASA. We thus divided the cathodes of the anode supported mt-SOFCs into segments as shown in Fig. 1. We hence call this method more specifically “electrode-segmentation”.

The resolution of the spatial characterization is dependent on the dimension of the segments. As the dimension of the segment reduces, the resolution increases. This in turn enlarges the total electronic isolation area among the segments. With the larger isolation area, the deviation among the measured and the intrinsic values grows. Moreover, the configuration of the peripheral equipment becomes practically more difficult. Considering all these parameters, we divided the cathode of the cell into three segments as shown in Fig. 1.

Longitudinal temperature, current, and impedance measurements.—The segment temperatures were sensed on the cathode surfaces by K-type thermocouples, separately. The segment currents were measured through three electrical loads (ELZ 175, Keisoku Giken Co. Ltd.), separately connected to the segments as depicted in Fig. 1. We carried out the electrical measurements by the four point probe method to eliminate the peripheral resistances. In this method, because the EASA is not divided into segments, there might be current flow in the lateral direction depending on the potential difference between the segments, that might affect the accuracy of the measurements. In SOFCs, however, the ceramic-based components enable the segmentation on the EASA. We thus divided the cathodes of the anode supported mt-SOFCs into segments as shown in Fig. 1.

Experimental

Spatial characterization method: electrode-segmentation.—In order to characterize the local properties that are spatially varying, the electrochemical active area (EASA) of a fuel cell can be divided into electronically isolated small partitions. We refer to these partitions hereafter as “segments”. The segments can be separately connected to thermocouples, electric loads, and frequency response analyzers (FRA), etc. for measuring the local temperature, current, and impedance, etc. By analyzing the longitudinal variation of these local properties, we can elaborate the concerning physical and electrochemical processes, such as ionic conductivity, kinetic limitations, and mass transport limitation, etc. We call this spatial characterization method “segmentation”. Especially in polymer electrode membrane fuel cells, the segmentation is usually realized on the gas distribution plates due to practical restrictions. In this method, because the EASA is not divided into segments, there might be current flow in the lateral direction depending on the potential difference between the segments, that might affect the accuracy of the measurements.

Figure 1. A picture of a cathode-segmented anode-supported microtubular SOFC accompanied with a schematic of the experimental setup.

Downloaded on 2018-07-20 to IP 207.241.231.82 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).
are averaged and plotted. The standard deviation for the current measurements is approximately 0.01 whereas it is circa 0.1 for the temperature measurements.

We measured the segment impedances at a DC voltage of 0.7 V to identify the variations under the realistic conditions. We used an NF 5022 FRA (NF Co. Ltd.) that swept the frequency from 100 kHz to 0.1 Hz for an AC perturbation voltage of 10 mV (pk-pk). With the same FRA and settings, we measured the segment impedances sequentially. Due to the instability between 100 kHz-10 kHz that presumably arose from the incompatibility among the FRA and the external electric load, we could not measure the ohmic resistances (high frequency resistance) of the segments. To measure the ohmic resistance, we employed a Solartron 1280Z FRA (Solartron Analytical) that ensured the compatibility between the FRA and its internal electric load (no external electric load).

**Fabrication of the cathode-segmented anode-supported micro-tubular SOFC.**—We preferred a commercial tubular anode substrate that is manufactured by Repton Co. Ltd. The substrate was composed of NiO/YSZ (65:35 wt%). Upon reduction, this substrate yields a porosity of ~37%. We dip-coated the substrates with 8YSZ electrolyte and then sintered it at ~1700 K for two hours. By masking the electronically-isolated areas for the electrode-segmentation, we brushed-coated the cathode slurry of LSM/YSZ (La0.7Sr0.3MnO3/YSZ,10:3 wt%) (Daiichi Kigamo Kagaka Kogyo Co. Ltd.) onto the electrolyte surface. We sintered the cathodes at ~1323 K for two hours, so that we obtained the cells resemble the one in Fig. 1. Moreover, we coated silver-paste onto the cathode (segment) surfaces to enhance the electronic conductivity. Finally, thermocouples and silver wires were connected for temperature, current, and impedance measurements.

**Cell operation conditions.**—Prior to the gas supply, the segment temperatures were risen to ~1073 K by an electric furnace depicted in Fig. 1. Though the cell was placed in a quartz tube to prevent the heat loss to the surrounding for maintaining a constant temperature along the cell, the segment temperatures slightly (±1 K) diverged, which is in the precision range of the thermocouples.21,22 The fuel inlet tube was preheated to avoid the convective cooling in the anode side. The fuel outlet tube was also heated to inhibit the condensation of the product water vapor. On the cathode side, the inlet air temperature was raised about 1048 K by the electric furnace before reaching the cathode surface. From this perspective, we can state that the air was preheated as well. The flow configuration was switched between co- and counter-flow by simply exchanging the fuel inlet and outlet, i.e., the air flow direction was kept the same, as Fig. 1 illustrates. Gas flow rates were metered at 298 K and 100 kPa with mass-flow-controllers (SEC-E40MK3, Horiba STEC) governed by LabVIEW 8.6. In order to reduce NiO to Ni, a dry mixture of H2/N2 (99.99% pure) was initially fed to the anode for two hours.

Although we have devoted this study to the fundamental understanding of the temperature variations, as pointed out in the previous paragraphs, we paid a particular attention on choosing the experimental conditions (furnace temperature, flow rates, pressure, etc.) appearing in the practical applications. Considering the design of a single mt-SOFC, our experimental setup also resembles that employed in reality. Despite we conduct this study on a single mt-SOFC, a number of mt-SOFCS are bundled for the real applications. Owing to the fact that the same processes involve in the temperature variations, this fundamental study sheds light on the bundles as well.

**Results and Discussion**

To be consistent throughout the analyses of the variations in the co- and counter-flow configurations, we call the segments “up-, mid-, and down-segments” referring to the positions depicted in Fig. 1. We plot the segment temperatures as “temperature rise (∆Tseg)” that is the difference between the segment temperature (Tseg) and the furnace temperature (Tfur) = 1073 K.

\[
\Delta T_{seg} = T_{seg} - T_{fur}
\]

**The prevailing process on the longitudinal temperature variations.**—Since the reversible and irreversible losses are essential to disclose the other involving processes. Therefore, we begin our analysis at a high fuel flow rate to keep the fuel utilization low (Uf = 29.3% at 0.4 V). Under these conditions with the co-flow configuration, the measured segment I-V curves are depicted in Fig. 2. In this figure, the I-V curves almost overlap, i.e., the current variations are quite small among the segments through the voltage range. Thereby, we expect insignificant temperature variations.

Fig. 3 provides sufficient evidence to justify our expectation for the longitudinal temperature variations. In this figure, all the segment temperatures are higher than the furnace temperature at open circuit voltage (OCV). The temperature rise at OCV stems from the combustion of the leaking hydrogen that mainly occurs around the sealant (Aremco, Ceramabond 552) among the cell and metal holders. The
extent of the hydrogen leakage can be estimated via analyzing the heat balance within the system. For this analysis, the initial state of the system right after the hydrogen and air supply was regarded; and the segment temperatures were in-situ measured as plotted in Fig. 4.

In the transition state, the total heat production rate $\Delta H_{\text{leak}}$ (kW) is released via the combustion of the leaking hydrogen $\Delta n_{\text{H2}}$ (mol/s)

$$\Delta H_{\text{leak}} = \Delta H_{\text{HOR}} \Delta n_{\text{H2}} \quad [8]$$

While $Q_{\text{cell}}$ (kJ) is absorbed by the cell, $Q_{\text{conv}}$ (kW) is simultaneously removed by the air flow via the convective heat transfer on the cathode surface. Owing to the relatively low heat conductivity of the sealant ($k = 30 \text{ W/mK}$), the conductive heat transfer to the adjacent piping is neglected. Since the segment surfaces are coated with silver-paste that resembles the gray body (poor radiative properties), the radiative heat transfer is ignored, too. In this regard, the heat balance within the system can be formulated as

$$\int_0^t \Delta H_{\text{HOR}} dt = Q_{\text{cell}} + \int_0^t Q_{\text{conv}} dt \quad [10]$$

Herein the time interval for the temperature rise in the segments $t = 140$ s from Fig. 4. When the $Q_{\text{cell}}$ and $Q_{\text{conv}}$ are calculated, $n_{\text{H2, leak}}$ (mol/s) can be found.

Due to the varying thermo-physical property of the cell components

$$Q_{\text{cell}} = Q_i + Q_{\text{el}} + Q_c \quad [11]$$

where the subscript “$i$” stands for the anode, “el” for the electrolyte, and “c” for the cathode. For

$$i \in (a, el, c) \quad Q_i = \rho V_i C_p i (T_i - T_0) \quad [12]$$

where $V$ (m$^3$) represents the solid volume of the cell. The density $\rho$ and the heat adsorption coefficient $C_p$ of the cell components are given as 3310, 5160, and 3030 (kg/m$^3$); and 450,470, and 430 (J/kgK) for Ni/YSZ (anode), 8YSZ (electrolyte), and LSM/YSZ (cathode), respectively. According to the Newton’s law

$$Q_{\text{conv}} = h A (T_{\text{cell}} - T_{\infty}) \quad [13]$$

where $A$ (m$^2$) is the surface area over which the convection takes place and the heat transfer coefficient $h = 2.8 \text{ W/m}^2\text{K}$ calculated from the Nusselt number

$$Nu = h D_h / k = 10.2 \quad [14]$$

which is accepted due to the rather low Reynolds number ($Re = 6.98 \times 10^6$). Herein, $D_h$ (m) is the hydraulic diameter and $k$ (W/mK) the heat conductivity of air. $T_{\infty} = 1048 \text{ K}$ is taken from Fig. 4. Considering the arithmetic average temperature of the segments at $t = 140$ s as

$$T_{\text{seg}} = (T_{\text{seg, up}} + T_{\text{seg, mid}} + T_{\text{seg, down}}) / 3 \quad [15]$$

The fuel leakage rate

$$\dot{n}_{\text{H2, leak}} / \dot{n}_{\text{H2, in}} \times 100 = 0.77 \quad [16]$$

Such amount of leakage rate is acceptable considering the operation at high temperature.

In Fig. 3 the slight temperature drops with the rising segment currents in the low current density region are associated with the reduction in the fuel leakage rate owing to the increasing consumption of hydrogen by the electrochemical reaction. Though the up- and down-segments are nearest to the main combustion areas (sealant), at OCV the down-segment exhibits the smallest temperature whereas the mid- and up-segments’ temperatures are similar and rather high. Since the cell is positioned in the geometrical centre of the furnace, and the segment temperatures were set approximately the same prior to the gas supply, we do not expect such variations from the radiative and conductive heat transfers upon supplying the gases. Thereby, we ascribe this temperature distribution profile to the convective heat transfer. In fact, this argument is verified by Fig. 5 wherein the temperature of the down-segment at 0.7 V significantly drops with the rising air flow velocity. Note that no mass transport limitation exists in the cathode side in this velocity domain.

As Fig. 1 displays, the convective heat transfer is occurring in two distinct interfaces: firstly, between the quartz tube and air, secondly, between air and the cell. The air is initially heated about 1048 K (Fig. 4) by the quartz tube which is in direct contact with the furnace. This heated air proceeds along the cell; and cools it down. Although the flow velocity range shown in Fig. 5 is quite small, forming laminar flow ($Re = 6.98$ at 2.0 cm/s), the thermal boundary layer is not developed within the entrance region of the cell. This means that the flow in the entrance regions is not laminar yet. As a result, the local Nusselt number/heat transfer coefficient within the entrance region of the cell is higher than what we assumed while estimating the hydrogen leakage rate and it is a function of the flow velocity. The entrance length $L_h$ (m) is given as

$$L_h = \frac{0.05 \text{Re} D_h}{[17]}$$

From Eq. 17, $L_h = 12.9 \text{ mm}$ can be calculated, being slightly longer than the length of the down-segment (9 mm). The down-segment’s temperature is thus highly affected by the air flow velocity. In this
Figure 6. I-V curves of the segments with the counter-flow configuration for H$_2$/N$_2$ = 80/120 cm$^3$/min and Air = 2000 cm$^3$/min.

Figure 7. I-T curves of the segments with the counter-flow configuration for H$_2$/N$_2$ = 80/120 cm$^3$/min and Air = 2000 cm$^3$/min.

Figure 8. Impact of the convective heat transfer on the segment temperatures at 0.7 V with the counter-flow configuration for H$_2$/N$_2$ = 80/120 cm$^3$/min. 2000 cm$^3$/min is equivalent to 2.0 cm/s.

Influence of the temperature variations on the ohmic resistance along the cell.—As mentioned previously, assuming that the main contribution to the ohmic resistance of an SOFC comes from the ionic resistance, Morel et al. proposed a method for predicting the temperature variations over a cell comprised a rather thick (500 μm) electrolyte by using the relationship between the ionic conductivity and temperature given by Eq. 6. In fact, the thickness of the electrolyte in our study is relatively smaller (~20 μm), and approximately constant along the cell. However, the significant temperature variation between the down- and up-segments depicted in Figs. 5 and 8 causes notable ohmic resistance difference between them. Table 1 presents
Table I. Impact of the temperature variations on the ohmic resistance along the cell for H\textsubscript{2}/N\textsubscript{2} = 80/120 cm\textsuperscript{3}/min. 2000 cm\textsuperscript{3}/min is equivalent to 2.0 cm/s.

| Air Flow Velocity (cm/s) | Co-Flow | Counter-Flow |
|-------------------------|---------|--------------|
| Up-Segment   | 0.127   | 0.125        |
| Down-Segment | 0.145   | 0.149        |

the resistances measured by impedance spectroscopy for the co- and counter-flow configurations.

At 1.0 cm/s, the ohmic resistance in the down-segment is ~16% higher than that in the up-segment with both flow configurations. As the air flow velocity increases, the down-segment’s temperature drops (Figs. 5 and 8), which results in higher local ohmic resistance. In contrast, the temperature of the up-segment is weakly dependent on the air flow velocity, thus, the ohmic resistance of this segment changes slightly. Consequently, the resistance difference along the cell rises. This implies that at a constant cell voltage, the down-segment suffers from both ohmic and kinetic limitations that influence both the current and temperature variations.

Longitudinal current variations arising from the nernst-loss coupled with the temperature variations.—While analyzing the impact of the convective heat transfer on the longitudinal temperature variations in the preceding subsection, high fuel flow rate (low fuel utilization) was opted to eliminate the contribution from the Nernst-loss. This analysis has disclosed the effect of the temperature variations on the current variations (counter-flow). However, we are aware of the fact that the Nernst-loss in the realistic fuel flow conditions is significant.\textsuperscript{12} Therefore, we will analyze the impacts of both temperature variations and Nernst-loss on the current variations under the realistic conditions in the following.

Fig. 9 illustrates the longitudinal current variations measured at the realistic fuel flow conditions with the co-flow configuration. In comparison to the high fuel flow rate, herein the fuel utilization is rather high (U\textsubscript{f} = 50% at 0.4 V). As a result, the current variations are larger, especially at lower voltages. Despite the higher temperature of the up-segment shown in Fig. 10 (mid- and up-segments have similar temperatures), the lower performances of the mid- and up-segments indicate that the Nernst-loss is limiting factor. On the other hand, the longitudinal current distribution profile is different, i.e., the up-segment exhibits better performance than the mid-segment. Taking merely the continuous hydrogen consumption into account, such a current distribution profile would not be acceptable. However, it is known that the increasing concentration of the product water favors the HOR to an extent.\textsuperscript{13,28} Relying on the development of a similar longitudinal temperature distribution profile with the counter-flow configuration owing to the prevailing convective heat transfer, we can analyze the interrelation among the temperature and current variations. We will carry out this analysis on Fig. 11 that presents the I-V curves measured under the realistic conditions with the counter-flow configurations. Herein, the current variations are quite large and they become larger with the rising fuel utilization (decreasing cell voltage). Beyond 0.5 V, the down-segment’s current reduces whereas the other segments’ currents rise; namely, the down-segment experiences severe fuel depletion. In comparison to the co-flow case (Fig. 9), the longitudinal current distribution profile is distinct as well. Since the only difference between Figs. 9 (co-flow) and 11 (counter-flow) is the fuel flow direction, we
can attribute the large current variations to the high temperature of the up-segment (Fig. 12) that boosts the local current production. Eventually, the Nernst-loss becomes more significant in the down-segment, changing the longitudinal current distribution profile.

Even though the longitudinal temperature and current variations at low fuel utilization conditions are analogous in terms of the flow configurations, they become rather different under the realistic operation conditions where the temperature and concentration variations couple. Through the analyses of Figs. 9–12, it is evident that the temperature and current variations are larger with the counter-flow configuration. The larger temperature and current gradients are in good agreement with the numerical studies which estimates quantitatively higher gradients indeed.6,15,17

The boost in the up-segment’s current production attributed to the high local temperature (Fig. 12) can be elaborated through the impedance analysis in a wide frequency range. In fact, we have already discussed the ohmic resistance of the up- and down-segments on Table I. We have concluded that the up-segment exhibits smaller ohmic resistance owing to its higher temperature. The effect of the high temperature on the other processes will be analyzed through longitudinal impedance variations.

Impedance spectra of the up- and down-segments measured with the counter-flow configuration are illustrated in Fig. 13. Though it is not easy to distinguish, we can define three different frequency ranges, ∼10 kHz − 376 Hz, ∼376 Hz − 4 Hz, and ∼4 Hz − 0.1 Hz, as the high frequency impedance (HFI), the medium frequency impedance (MFI), and the low frequency impedance (LFI), respectively. Herein, the significant impedance difference in the MFI and LFI of the segments is obvious. This notable impedance difference confirms the I-V curves depicted in Fig. 11. Namely, the total impedance is remarkably smaller in the up-segment that operates at high temperature (Fig. 12) and receives hydrogen at the highest concentration. Owing to the small impedance, the up-segment’s current is boosted, so that the Nernst-loss rises toward the down-segment. As a result, the down-segment does not only suffer from the Nernst-loss, but also from the rather low temperature difference. As the up-segment operates at higher temperature (Figs. 10 and 12), the MFI and LFI in this segment are smaller. Consequently, the current in the up-segment is boosted, resulting in larger Nernst-loss in the down-segment and eventually increasing the current variations (Fig. 11). In contrast, the down-segment operates at lower temperature that raises the MFI and thus reduces the impedance of this segment is smaller than that of the down-segment owing to the higher temperature and restricted Nernst-loss. Namely, the current distributions are levelled by the temperature variations. As a result, the further temperature variations stemming from the current variations are mitigated.

Comparison of the “upstream” segments in the co- and counter-flow configurations supports the coupling of the temperature and current variations. Herein the “upstream” segment refers to the segment that receives the highest hydrogen concentration, i.e., the up-segment in the counter-flow case (Fig. 13) whereas the down-segment in the co-flow case (Fig. 14). Fig. 15 plots the impedance spectra of the upstream segments in both flow configurations. In this figure, the difference between the impedances of the up- and down-segments comes from the temperature difference. As the up-segment operates at higher temperature (Figs. 10 and 12), the MFI and LFI in this segment are smaller. Consequently, the current in the up-segment is boosted, resulting in larger Nernst-loss in the down-segment and eventually increasing the current variations (Fig. 11).
Current variations and the convective heat transfer, we conducted the investigation of the processes involving in the temperature variations in a segment.

Down-segment’s temperature drops (Fig. 12), which in turn increases the local temperature is clearly shown. As the air flow velocity rises, the impedance spectra of various air flow velocity with the counter-flow configuration as plotted in Fig. 14. Ultimately, the longitudinal current variations become smaller (Fig. 9).

Nernst-loss. Ultimately, the longitudinal current variations become smaller (Fig. 9).

Since the activation and mass transport (including the Nernst-loss) impedances are coupled in the previous impedance analyses, the sensitivity of the local impedance to the temperature cannot be observed evidently. Yet, due to the direct impact of the convective heat transfer on the down-segment’s temperature, it is possible to control the temperature of this segment at the same hydrogen concentration owing to the almost constant currents in the mid- and up-segments for identifying the effect of the local temperature on the local impedance. With this intention, we measured the impedance spectra of the down-segment measured at 0.7 V with the counter-flow configuration for H2/N2 = 40/40 cm3/min. Air = 2000 cm3/min equivalent to 2.0 cm/s.

Conclusions

Aiming the mitigation of the longitudinal temperature variations that give rise to the thermal stresses, we investigated the contribution of the processes involving in the temperature variations in a microtubular solid oxide fuel cell. Giving particular emphasis on the current variations and the convective heat transfer, we conducted the analyses on the properties which we in-situ obtained by the electrode-segmentation method for the co- and counter-flow configurations. In order to disclose the impact of the temperature variations on the current variations, we elaborated the interrelationship among the temperature, current, and impedance variations. Through these analyses, we reach the following conclusions:

1. Air flow at excess rates dominate on the longitudinal temperature variations causing a remarkable temperature gradient in the air entrance region, that poses high risk of mechanical failure.
2. In terms of the flow configuration, counter-flow exhibits larger temperature, current, and impedance variations than the co-flow configuration.
3. Although the current variations are reduced in the co-flow configuration, they are enhanced in the counter-flow configuration by the temperature variations.
4. Despite the preheating, the air flow velocity exhibits substantial impact on the longitudinal temperature distribution and thus on the current variations. We hence anticipate that the reduction of the excess air flow would alleviate the variations and the associated thermal stresses.

Acknowledgment

The first author provides his gratitude to the Japanese Government for the MEXT Super Global Scholarship. The authors acknowledge the JSPS (Japanese Society for Promotion of Science) for Grant-in-Aid for Young Scientists (B) 25820064.

References

1. K. Fischer and J. R. Seume, “Impact of the temperature profile on thermal stress in a tubular solid oxide fuel cell,” J. Fuel Cell Sci. Technol., 6, 011017 (2009).
2. A. Selimovic, M. Kemm, T. Torisson, and M. Assadi, “Steady state and transient stress analysis in planar solid oxide fuel cells,” J. Power Sources, 145, 465 (2005).
3. O. Rachedi, I. Wärendorff, and M. Assadi, “Experimental investigation of temperature distribution over a planar solid oxide fuel cell,” Applied Energy, 105, 155 (2013).
4. A. Nakajo, C. Stiller, G. Harkgard, and O. Bolland, “Modelling of thermal stresses and probability of survival of tubular SOFC,” J. Power Sources, 158, 287 (2006).
5. C. M. Dikwal, W. Bujalski, and K. Kendall, “The effect of temperature gradients on thermal cycling and isothermal ageing of micro-tubular solid oxide fuel cells,” J. Power Sources, 193, 241 (2009).
6. H. Yakabe, T. Ogawa, M. Hishimura, and J. Yasuda, “3-D model calculation for planar SOFC,” J. Power Sources, 102, 144 (2001).
7. C.-K. Lin, T.-T. Chen, Y.-P. Chyou, and L.-K. Chiang, “Thermal stress analysis in a planar SOFC stack,” J. Power Sources, 164, 238 (2007).
8. A. A. Kulikovsky, “Temperature and current distribution along the air channel in planar SOFC stack: model and asymptotic solution,” J. Fuel Cell Sci. Technol., 7, 011015 (2010).
9. B. Morel, R. Roberge, S. Savoie, T. W. Napporn, and M. Meunier, “An experimental evaluation of the temperature gradient in solid oxide fuel cells,” Electrochim. Solid-State Lett., 10, B31 (2007).

Figure 15. Impedance spectra of the “upstream” segments in the co-flow (down-segment) and counter-flow (up-segment) configurations at 0.7 V for H2/N2 = 40/40 cm3/min and Air = 2000 cm3/min equivalent to 2.0 cm/s.

Figure 16. Impact of the temperature drop achieved by increasing air flow velocity on the impedance of the down-segment measured at 0.7 V with the counter-flow configuration for H2/N2 = 40/40 cm3/min. Air = 2000 cm3/min equivalent to 2.0 cm/s.
18. A. A. Kulikovsky, “A simple equation for temperature gradient in a planar SOFC stack,” Int. J. Hydrogen Energy, 35, 308 (2010).
19. L.-K. Chiang, H.-C. Liu, Y.-H. Shiu, C.-H. Lee, and R.-Y. Lee, “Thermo-electrochemical and thermal stress analysis for an anode-supported SOFC cell,” Renewable Energy, 33, 2580 (2008).
20. M. G. Santarelli, P. Leone, M. Cali, and G. Orsello, “Experimental analysis of the voltage and temperature behaviour of a solid oxide fuel cell generator,” J. Fuel Cell Sci. Technol., 4, 143 (2007).
21. http://www.maximintegrated.com/en/app-notes/index.mvp/id/5032, last visited on 13th August, 2015.
22. http://www.omega.com/temperature/z/pdf/z019-020.pdf, last visited on 13th August, 2015.
23. K. Fischer and J. R. Seume, “Location and magnitude of heat sources in solid oxide fuel cells,” J. Fuel Cell Sci. Technol., 6, 011002 (2009).
24. P. Auerkari, “Mechanical and physical properties of engineering alumina ceramics,” VTT Manufacturing Technology, Research Notes 1792.
25. Y. A. Cengel, “Internal Forced Convection,” in: Y. A. Cengel, Heat Transfer, A Practical Approach, 2nd Edition, McGraw-Hill, 2002, p. 424.
26. V. M. Janardhanan and O. Deutschmann, “Numerical study of mass and heat transport in solid-oxide fuel cells running on humidified methane,” Chem. Eng. Sci., 62, 5473 (2007).
27. W. M. Kays and H. C. Perkins, in Handbook of Heat Transfer, 2nd ed., W. M. Rohsenow, J. P. Hartnett, and E. N. Ganic, Editors, McGraw-Hill, New York (1985).
28. S. H. Jensen, A. Hauch, P. V. Hendriksen, M. Mogensen, N. Bonanos, and T. Jacobsen, “A method to separate process contributions in impedance spectra by variation of test conditions,” J. Electrochem. Soc., 154, B1325 (2007).