Exploring the effects of symmetrical and asymmetrical relative humidity on the performance of H$_2$/air PEM fuel cell at different temperatures

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Abstract

This article is dedicated to study the interlinked effects of symmetric relative humidity (RH), and asymmetric RH on the performance of H$_2$/air PEM fuel cell at different temperatures. The symmetric and asymmetric RH were achieved by setting the cathode relative humidity (RHC) and anode relative humidity (RHA) as equal and unequal values, respectively. The cell performance was evaluated by collecting polarization curves of the cell at different RH, RHC and RHA and at different cell temperatures (T_{cell}). The polarization curves along with the measured internal cell resistance (membrane resistance) were discussed in the light of the present fuel cell theory. The results showed that symmetric relative humidity has different impacts depending on the cell temperature. While at RH of 35% the cell can show considerable performance at $T_{cell} = 70\,°C$, it is not so at $T_{cell} = 90\,°C$. At $T_{cell} = 70\,°C$, the cell potential increases with RH at lower and medium current densities but decreases with RH at higher currents. This was attributed to the different controlling processes at higher and lower current densities. This trend at 70 °C is completely destroyed at 90 °C. Operating our PEM fuel cell at dry H$_2$ gas conditions (RHA = 0%) is not detrimental as operating the cell at dry Air (O$_2$) conditions (RHC = 0%). At RHA = 0% and humidified air, water transport by back diffusion from the cathode to the anode at the employed experimental conditions can support reasonable rehydration of the membrane and catalysts. At RHA = 0, a possible minimum RHC for considerable cell operation is temperature dependent. At RHC = 0 conditions, the cell can operate only at RHA = 100% with a loss that depends on T_{cell}. It was found that the internal cell resistance depends on RH, RHA, RHC and T_{cell} and it is consistent with the observed cell performance.
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1. Introduction

Proton exchange membrane fuel cells (PEMFC) have gained a growing interest in recent years due to its possible replacement for traditional engines, primarily because of their high power density and zero emission features [1–3]. Despite the significant progress in the PEM fuel cell technology over the past decade, high performance with proper stability and reliability as well as low cost are yet to be achieved before fuel cells can replace the current ordinary power generators. Water management rep-

resents one of the main critical and design issues of PEMFCs because the membrane's hydration in a PEMFC determines the performance and durability of a PEMFC. If the membrane is not properly hydrated, it exhibits higher ionic resistance and can even be irreversibly damaged in some extreme cases. Polymer membrane materials used in PEMFCs should be hydrated in order to maintain high proton conductivity. Meanwhile, excess water must be removed to prevent flooding. Membrane hydration is affected by the water transport phenomena in the membrane itself, which in turn is affected by the condition of the inlet gases and the operating parameters of the fuel cell [4]. An optimum water balance during the operation of PEMFCs is always a main goal to achieve higher performance. The mechanism of water transport inside the membrane can be; electro-osmotic drag (water molecules are dragged by the protons traveling from the anode to the cathode), back diffusion (water is transferred

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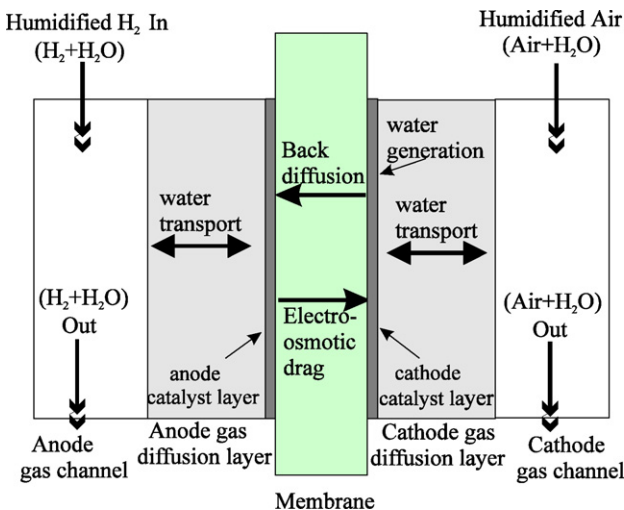
Fig. 1. Schematic of the water transport within a H_2/air PEM fuel cell.

into the membrane due to the water concentration gradient from the cathode to the anode), and convection (due to pressure gradients between the cathode and anode). However, the convection effect is generally negligible compared to the effects of electro-osmotic drag and the back diffusion, due to a very low membrane hydraulic permeability [4]. Fig. 1 shows a scheme of the water transport process in a typical H_2/air PEM fuel cell. The two main water transport processes in the membrane are electro-osmotic drag and back diffusion; these affect the water balance in a PEMFC and determine the membrane hydration. Optimum water balance between the anode and cathode is crucial for achieving good cell performance because the water level in a fuel cell strongly affects not only the membrane properties, but also reactant transport and electrode reaction kinetics [4].

Mathematical models have been developed to account for the effects of relative humidity and temperature on the performance and water transport of PEM fuel cells [5–11]. For instance, Bernardi and Verbrugge [5–7] developed a model to determine fuel cell operating conditions that would result in optimal balance between water that is formed in the fuel cell reaction and that must be removed to prevent flooding. The modeling results revealed that humidification of reactant gases could be adjusted as the current density was varied to accommodate the changing demand for water in the fuel cell. The analytical results also identified the conditions under which reactant transport limitations govern the behavior of the fuel cell. Yi and Nguyen [8] developed an along-the-channel model for evaluating the effects of various design and operating parameters on the performance of a proton exchange membrane (PEM) fuel cell. Their results showed that the performance of a PEM fuel cell could be improved by anode humidification and positive differential pressure between the cathode and anode to increase the back transport rate of water across the membrane. Nguyen and White [12] developed a water and heat management model for fuel cell and used the model to investigate the effectiveness of various humidification designs. They found that, at high current density ($>1\,A\,cm^{-2}$), ohmic loss in the membrane is responsible for a large fraction of the voltage loss in the cell, and back diffusion of water from

the cathode side of the membrane was insufficient to keep the membrane hydrated (i.e., conductive). Consequently, to minimize the ohmic loss, the anode stream must be humidified, and when air is used instead of pure O_2, the cathode stream must also be humidified. Okada et al. [13,14] used a linear transport equation based on the diffusion of water and the electro-osmotic drag to analyze water concentration profiles in the membrane.

While most PEMFC studies were performed at high relative humidity conditions (symmetric, i.e., with equal high RH at both cathode and anode) at various temperatures and pressures, the number of studies regarding the experimental investigation of the effects of operating the H_2 PEM fuel cell at dry H_2 gas conditions is considerable [15–23]. In the above studies non-zero values of RHA and RHC were used. However, studying the cell performance at either (RHA = 0 and RHC $\neq$ 0) and (RHA $\neq$ 0 and RHC = 0) is limited. The latter conditions can help to study the impacts of electro-osmosis and back diffusion of water on the rehydration of the membrane and the cell performance at variable conditions.

The aim of the present study is to explore the effects of the symmetric RH, and asymmetric RH (different values of RHA and RHC) on the performance of the H_2/air PEM fuel cell at different temperatures. Polarization curves are collected at different conditions and discussed along with the internal cell resistance (membrane resistance). The influence of the electro-osmosis and back diffusion may be illustrated under some experimental conditions.

2. Experimental

The membrane electrode assembly (MEA) was purchased from Umicore Fuel cells (Germany). Nafion 212 membrane and $1.0\,mg\,cm^{-2}$ Pt catalyst, $1.0\,mg\,cm^{-2}$ Pt-Ru (0.766 Pt and 0.234 Ru) catalyst were used in all the experiments. The fuel cell of a JARI Standard type was purchased from NF Co. (Japan) and the MEA was of $25\,cm^2$ area.

The fuel cell overall performance was evaluated at different cell temperatures and at different relative humidity. In a set of experiments, the RH of the H_2 gas (anode compartment), RHA was kept constant = 0 and the relative humidity of air (O_2) (cathode compartment), RHC was changed from 0 to 100%. The reverse was done for air, i.e., keeping air dry and changing the relative humidity of H_2 (RHA) from 0 to 100%. The above setup may be called asymmetric relative humidity since the RH at the cathode and anode is different. In both cases the polarization curves of the cell were recorded along with the internal resistance of the cell. The flow field pattern was of serpentine type with opposite gas flow directions. The gas flow rate was kept constant during the whole course of measurements, i.e., at 300 and $1000\,cm^3\,min^{-1}$ for H_2 and air, respectively. These flow rates satisfy a ratio slightly higher than the stoichiometric ratio of H_2 and O_2 gases. The gas pressure was kept constant accordingly at 1.5 and 25.3 kPa for H_2 and air, respectively. Temperatures of the anode and cathode gas lines between the humidifiers and the cell were maintained at $20\,°C$ higher than the cell temperature to ensure no water condensation.
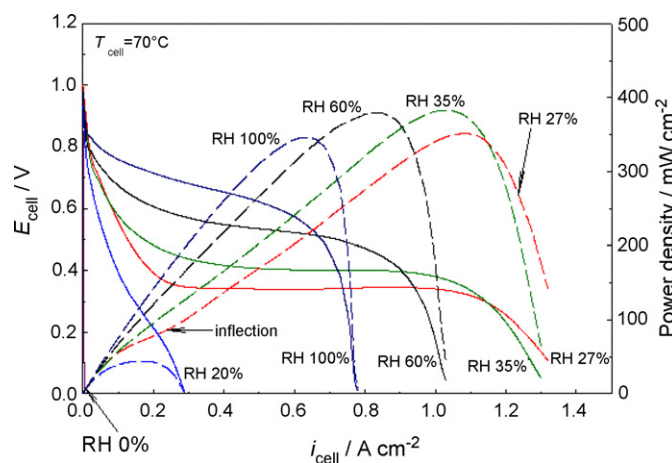
Fig. 2. Polarization curves and power density of the H$_2$/air PEM fuel cell at different RH (symmetric) at $T_{cell} = 70\,^\circ$C.

Fig. 3. Polarization curves and power density of the H$_2$/air PEM fuel cell at different RH (symmetric) at $T_{cell} = 90\,^\circ$C.

The polarization curves were measured by NF-Fuel Cell Test System (MTB-36714, Japan) coupled with a potentiostat. They were recorded using galvanodynamic technique with a current scan rate of 0.05 A min^{-1}. This scan rate was found to be lower enough to obtain stable constant values of potential. The current was scanned from lower currents (0 A) to higher positive values. The maximum current is taken where the cell voltage goes below a minimum value. This minimum depends on the operating conditions. The polarization curves were measured with the air electrode as the working electrode and the hydrogen electrode as the reference and counter electrode. Cell internal resistance was simultaneously measured with the cell potential at the different current densities using current interrupt technique. The cell internal resistance compromises mostly ionic resistance of the cell membrane and pure electrical (bulk and contact) resistances. By sufficient compression of the gas diffusion layer of the MEA, the pure and contact resistances can be considered as negligible. In this case the measured internal resistance can be considered as the membrane ionic resistance [24]. The difference in the total thickness of the seal gaskets and the total thickness of the MEA was 0.3 mm, which can be considered to be sufficient to neglect contact resistances [24].

3. Results and discussion

3.1. Polarization curves

In this section the relative humidity at both the anode and cathode is set at the same value (i.e., symmetric RH). Polarizations curves at different RH were collected at cell temperature, T_{cell} of 70 and 90 $^\circ$C as shown in Figs. 2 and 3, respectively. As the figures reveal, the effects of RH depend on the cell temperature. While RH of 27% can still support the cell performance at $T_{cell} = 70\,^\circ$C, it provides poor performance at $T_{cell} = 90\,^\circ$C. At $T_{cell} = 70\,^\circ$C, the cell performance is improved with higher RH at current density, $i_{cell} \leq 0.65$ A cm^{-2} but it is better at lower RH of 27, 35 and 60% at $i_{cell} > 0.65$ A cm^{-2}. At RH lower than 27% lower performance were obtained at this temperature (i.e.,

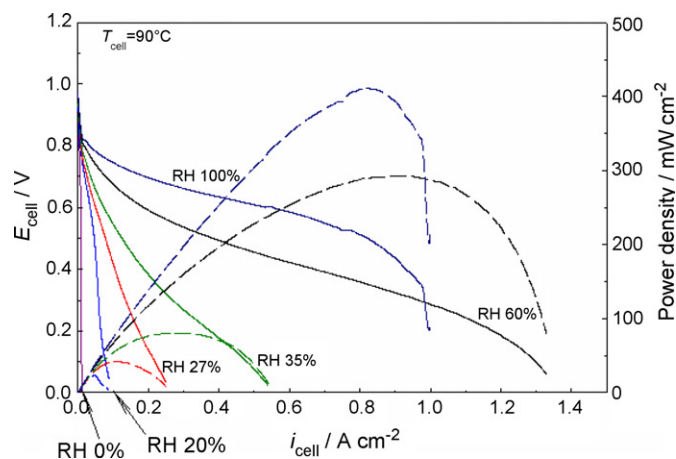$T_{cell} = 70\,^\circ$C). The minimum RH that keeps fair performance of the cell increases with the cell temperature. For instance, it is 27 and 60% at T_{cell} of 70 and 90 $^\circ$C, respectively. The cell performance is affected by the combined effects of RH and T_{cell}. Generally, at higher current densities the performance is limited by mass transfer resistance. This is evident from the lower performance of the cell at RH 100% than at RH of 27, 35 or 60% at 70 $^\circ$C. At lower and medium current density, however, the cell performance is controlled by the kinetic and ohmic resistance in favor of higher RH. This is evident from the high performance of the cell at RH = 100% at lower and medium current density. An interesting remark here is to observe the light inflection in the power density curve in Fig. 2 for the case of RH = 27% (the arrow in Fig. 2 points to this inflection). This was attributed to the self healing (rehydration) of the cell performance where at this current density water is produced at higher rates at the cathode causing higher back diffusion rates and hence better humidification of the cell which is required at this lower RH value. Note that under any T_{cell} and at RH = 0% (dry conditions) at both compartments the cell performance is destroyed due to highly dry membrane and catalyst layer. As T_{cell} increases the cell performance is improved only when RH = 100%. The kinetics of the reactions (especially at the cathode) and the conductivity of the membrane are improved with temperature. Note that at $T_{cell} = 90\,^\circ$C, however, the relative humidity controls the cell performance. Fig. 4 shows the potential as a function of RH at low (0.2 A cm^{-2}) and high (0.76 A cm^{-2}) current density at $T_{cell} = 70\,^\circ$C. At lower current density the potential increases with RH but at higher current density, it increases up to RH = 60% and then decreases at RH = 100%. Mass transfer resistance (especially at the cathode) is a controlling process at this situation. This trend is modified at much higher current density ($i_{cell} > 0.8$ A cm^{-2}). The above trend at $T_{cell} = 70\,^\circ$C is destroyed at 90 $^\circ$C and yet the cell performance increases with RH. At T_{cell} 90 $^\circ$C, the dry-out condition becomes a major problem and lower performances were obtained at lower RH. In this case RH is a more controlling factor than T_{cell}.

The above results were collected by using symmetric RH at both anode (RHA) and cathode (RHC). Studying the effects
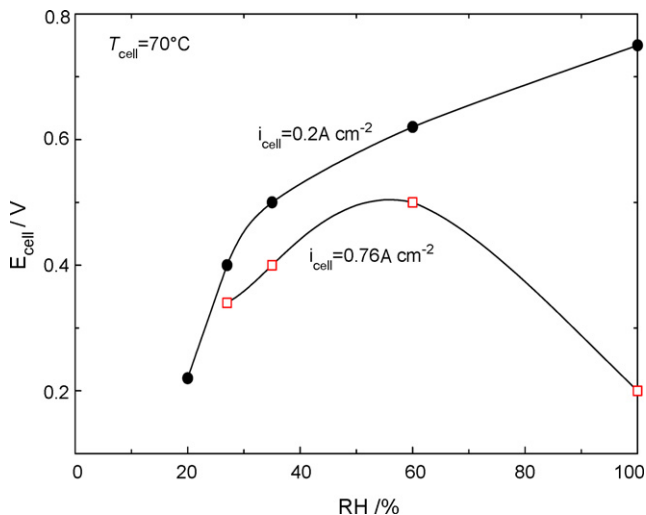
Fig. 4. E_{cell} as a function of RH at different i_{cell} and $T_{cell} = 70\,^{\circ}C$.
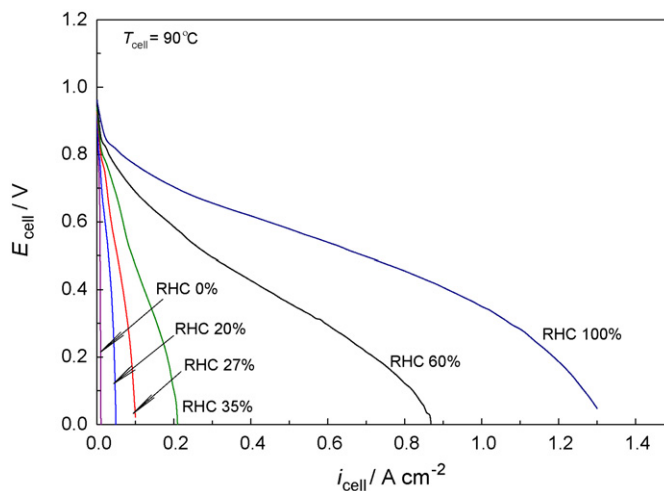

Fig. 6. Polarization curves of the H_2/air PEM fuel cell at different RHC (asymmetric) and RHA = 0% at $T_{cell} = 90\,^{\circ}C$.

of asymmetric RH (different RHA and RHC) on the performance of the cell may help to discuss the rehydarion capability of the membrane at different combinations of RHA and RHC. It may be possible to recognize and evaluate the impacts of electro-osmosis and back diffusion on the rehydration process. The effects of RH of the individual compartments; i.e., the cathode (air) and the anode (H_2 gas) are discussed here. Meanwhile we may answer these questions; is it possible to operate the fuel cell at considerable performance while one of the compartments suffers from dry conditions and the other is humidified? What is the minimum RH on the humidified compartment which can sustain the cell performance? This can be answered under the present experimental conditions.

Figs. 5 and 6 show the effects of the relative humidity of the cathode, RHC at a fixed condition of dry anode compartment (RHA = 0) at $T_{cell} = 70$ and $90\,^{\circ}C$, respectively, on the cell performance. At RHA = 0 and variable RHC, the cell can sustain reasonable performance at a specific minimum RHC dependent
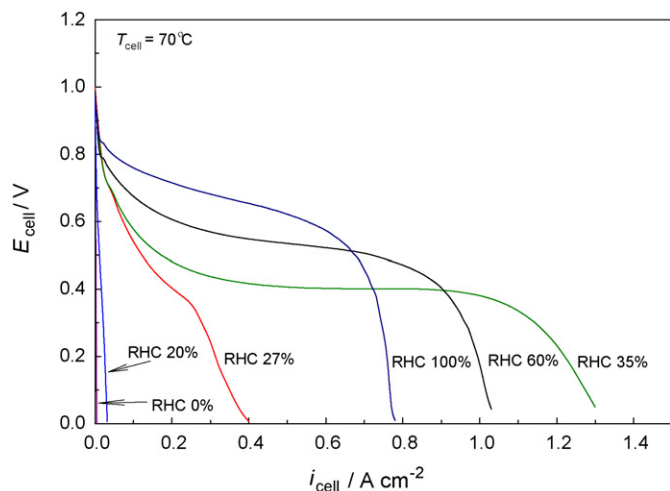

Fig. 5. Polarization curves of the H_2/air PEM fuel cell at different RHC (asymmetric) and RHA = 0% at $T_{cell} = 70\,^{\circ}C$.

of the cell temperature. At the same T_{cell}, the cell performance in Figs. 5 and 6 (asymmetric case) is lost at relatively higher RHC values compared to the corresponding RH values used in Figs. 2 and 3 (i.e., symmetric case). Compare Figs. 2 and 5 at RH = 27% and RHC = 27%, respectively, at $T_{cell} = 70\,^{\circ}C$. As the cell temperature increases, the cell looses its performance at higher RHC values. The cell performance is lost at RHC = 27 and 60% at T_{cell} equals 70 and $90\,^{\circ}C$, respectively. These RHC values are higher than the symmetric humidity (RH) values at which the cell performance is lost as shown in Figs. 2 and 3. The fact that the cell performance is sustained at RHA = 0 and at specific values of RHC is attributed to the rehydration of the anode side of the Nafion membrane by back diffusion of water from the cathode to the anode. The difference in pressures (and flow rates) between the cathode and the anode (see the Section 2) helped in the water transport from the cathode to the anode. Also, oxygen reduction reaction is affected by mass transfer resistance more than H_2 oxidation reaction as evident from the larger size of the O_2 molecule compared to H_2 molecule.

A different situation arose when RHC was set to zero and RHA was varied from 0 to 100% at different temperatures. This is depicted in Figs. 7 and 8 at T_{cell} of 70 and $90\,^{\circ}C$, respectively. The cell performance is lost if RHC is kept 0% (dry cathode) and the anode compartment is humidified (i.e., RHA = values between 0 and 100%). Only at RHA = 100% at which the cell can keep moderate performance and this also depends on the cell temperature. For instance, the cell performance at $70\,^{\circ}C$ is better than at $90\,^{\circ}C$. This can be understood by considering the dryness of the Nafion membrane at higher cell temperatures. Electro-osmotic drag of water from the anode to the cathode is important only when the anode compartment humidity, RHA equals 100% albeit of a degree depends on T_{cell}. In general back diffusion of water from cathode to anode (when RHA = 0 and RHC $\neq$ 0) at a pressure gradient can support more reasonable operation conditions than electro-osmosis from anode to cathode (when RHC = 0 and RHA $\neq$ 0).
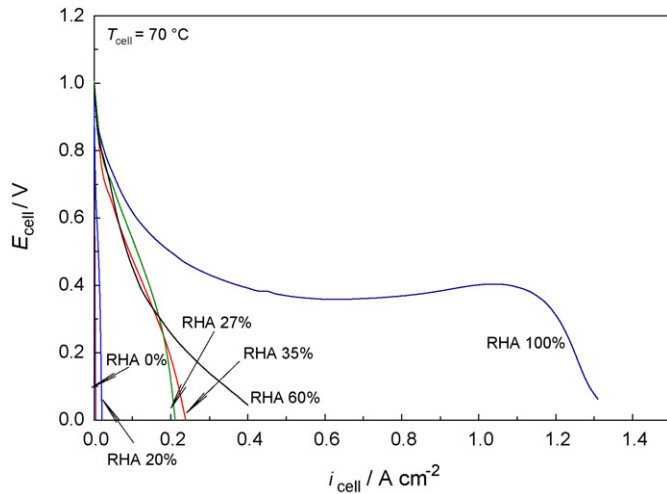
Fig. 7. Polarization curves of the H_2/air PEM fuel cell at different RHA (asymmetric) and RHC = 0% at T_{cell} = 70 °C.

3.2. Internal cell resistance

The internal resistance under the present conditions is a measure of the membrane ionic conductivity. A dryness of the membrane corresponds to higher values of the internal resistance. A discussion of plots of the membrane resistance versus the current density may help to understand the cell overall performance shown in the previous section. Fig. 9 shows the internal resistance as a function of the current density at different symmetric RH at T_{cell} = 70 °C. Only at RH ≥ 20%, the membrane resistance has limited measurable values and can be plotted as shown in Fig. 9. The values of the internal resistance at RH = 0% have much higher values and are not shown here. The internal resistance values that are plotted here are those at i_{cell} ≥ 0.1 A cm^{-2} to avoid the errors accompanying measurements at lower i_{cell} [24]. As the cell current increases, the resistance decreases to lower values depending on RH. This was attributed to the production of water at higher current densities. Back diffusion is possible at the present positive pressure gra-
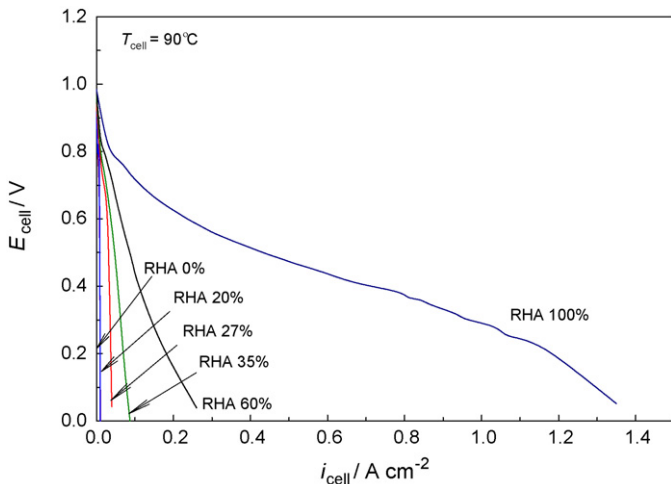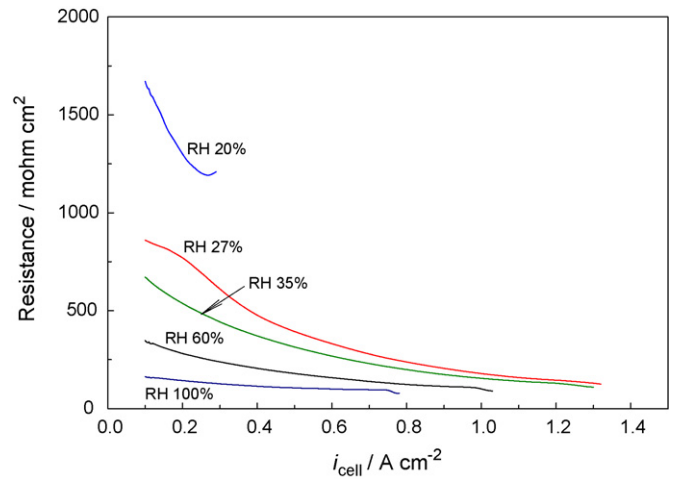

Fig. 9. Internal resistance (membrane resistance) of the H_2/air PEM fuel cell at different RH (symmetric) at T_{cell} = 70 °C.

dient from the anode to the cathode. Note that the decrease in the internal resistance (membrane ionic resistance) is steeper at lower RH than at higher RH. At higher RH the membrane is already well hydrated and further increase of the current density does not change it dramatically. At RH of 20%, the membrane is relatively dry and further production of water significantly decreases the resistance of the membrane. Similar results were obtained at T_{cell} = 90 °C and similar features and conclusion were found. At higher T_{cell}, however, there is a positive shift of RH at which dry-out condition occurs. It is ≤20, and <60% for T_{cell} 70 and 90 °C, respectively. This trend is in agreement with the general performance obtained in Figs. 2 and 3 at the same conditions.

The effects of RHC at dry anode (RHA = 0) can help to understand the performance of the cell shown in Figs. 5 and 6. Fig. 10 shows the effects of RHC at T_{cell} = 70 °C on the membrane resistance at different cell current and at RHA = 0. Only at RHC ≥ 27%, the membrane resistance has limited measurable values and can be plotted as shown in Fig. 10. The plots of the internal resistance at RHC 35, 60 and 100% have
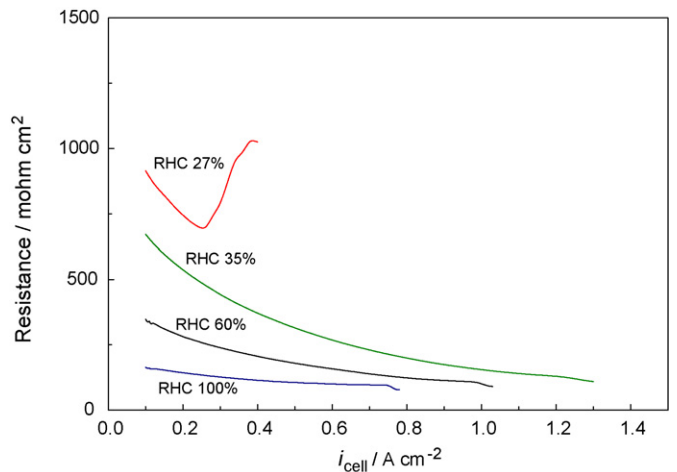

Fig. 8. Polarization curves of the H_2/air PEM fuel cell at different RHA (asymmetric) and RHC = 0% at T_{cell} = 90 °C.

Fig. 10. Internal resistance (membrane resistance) of the H_2/air PEM fuel cell at different RHC (asymmetric) and RHA = 0% at T_{cell} = 70 °C.
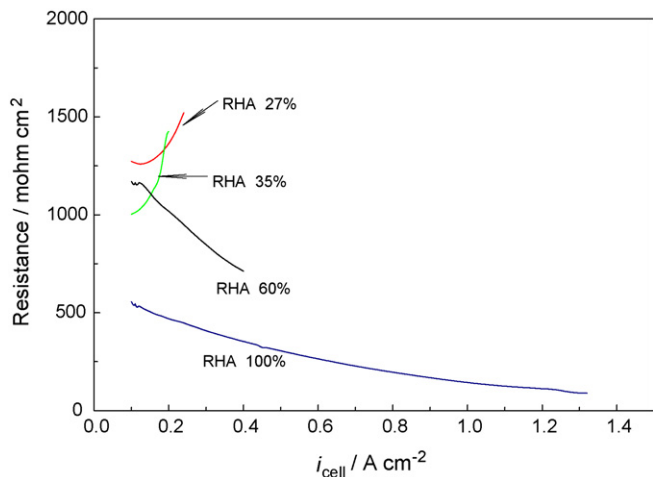
Fig. 11. Internal resistance (membrane resistance) of the H_2/air PEM fuel cell at different RHA (asymmetric) and RHC = 0% at $T_{cell} = 70\,°C$.

similar features as those at the corresponding RH shown in Fig. 9 (i.e., at RH = 35, 60 and 100%). At RHC = 27%, the membrane resistance decreases with increasing the current density to ca. 700 mΩ cm^2 and then begins to increase again at $i_{cell} = 0.27$ A cm^{-2}. This can be assigned to the dryness of the anode side of the membrane [24–26]. At higher cell currents and at lower RHC, the back diffusion of water from the cathode to the anode is not sufficient to support a considerable humidification of the membrane and consequently to affect the anode reaction (H_2 oxidation). At RHC = 27% and RHA = 0, the membrane resistance increases significantly at $i_{cell} > 0.27$ A cm^{-2}. This stiff increase in the cell resistance with the current increase indicates dry-out conditions. Similar results and conclusions were found at $T_{cell} = 90\,°C$ but with shifts in the relative humidity to higher values.

The effects of RHA at dry cathode (RHC = 0%) on the membrane resistance are shown in Fig. 11. Only at RHA ≥ 27%, the membrane resistance has limited measurable values and can be plotted as shown in Fig. 11. At RHA = 60 and 100% the membrane resistance shows the same behavior as in Figs. 9 and 10. However, the differences in the resistance at RHA = 60% and that at RHA = 100% are much higher than those obtained at the corresponding RH (Fig. 9) or RHC (Fig. 10). The increase of resistance with the current density at RHA = 27 is much steeper than that at RHC = 27% in Fig. 10. In general, the membrane resistance increases in the order of RHA > RHC > RH. The operation at RHC = 0% is detrimental under the employed experimental conditions. In the case of dry cathode electro-osmosis can support water only at higher RHA; the membrane dryness is evident at RHA < 60%. This trend is consistent with the performance of the cell discussed in the previous section.

The increase in cell resistance with current density is caused by the water distribution in the membrane due to the interplay of electro-osmotic drag transporting water toward the cathode and the back transport of water to the anode [24–27]. During fuel cell operation, with the proton transported from the anode to the cathode, water can be dragged by forming H_3O^+ and moved from the cell anode to the cathode. On the other hand, the cell cathode

has a higher water concentration, which drives water diffusion from the cathode to the anode due to a concentration gradient and capillary forces. The amount of water transported by electro-osmotic drag and back diffusion is dependent on current density and membrane properties and thickness. With increasing current density, the increased proton flux carries a large amount of water to the cathode. At very high current densities, the water back diffusion flux from the cathode to the anode is not sufficient to make up for the water lost at the cell anode, so the anode side of the membrane dries out causing a large membrane resistance. According to Zawodzinski et al. [25] and Buchi et al. [26], the back diffusion flux of water is directly proportional to the water diffusion coefficient or the water permeability of the membrane, which is determined by the local water content in the membrane.

4. Summary and conclusions

The combined effects of relative humidity (RH) of the cathode and anode in both symmetric and asymmetric arrangements on the performance of H_2/air PEM fuel cell were explored. The impacts of symmetric RH (i.e., the RH values at both cathode and anode are equal) on the cell performance depend on the cell temperature. While lower RH can support reasonable operation of the H_2/air PEMFC at lower cell temperature, higher RH should be applied to obtain good performance at higher cell temperature. The effects of asymmetric RH were studied by holding the RH of the cathode (RHC) or the anode (RHA) at 0% and changing the other one from 0–100%. While the cell can still operate at dry H_2 conditions (RHA = 0%) and RHC from 27–100% (dependent on T_{cell}), it can sustain its operation at dry air conditions (RHC = 0%) only at RHA = 100%. The internal resistance of the cell (the membrane resistance) reflects these phenomena at different conditions. Rehydration of the membrane was possible at specific conditions dependent on T_{cell}. For example, at $T_{cell} = 70\,°C$, these conditions were RH ≥ 27% (symmetric), (RHA = 100%, RHC = 0%) and (RHA = 0 and RHC ≥ 27%). These values were shifted to higher RH values at higher T_{cell}. The increase of the membrane resistance with current density at some conditions was assigned to dry-out condition. The dry-out conditions were obtained at specific values of RH, RHA, RHC highly dependent of the cell temperature.

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