



Effect of polytetrafluoroethylene-treatment and microporous layer-coating on the electrical conductivity of gas diffusion layers used in proton exchange membrane fuel cells

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ABSTRACT

The purpose of this study is to investigate the effect of polytetrafluoroethylene (PTFE)-treatment and microporous layer (MPL)-coating on the electrical conductivity of gas diffusion layers (GDLs), as used in proton exchange membrane fuel cells (PEMFCs). The results show that, for PTFE-treated GDLs, the electrical conductivity in orthogonal in-plane directions is almost invariant with the PTFE loading. On the other hand, the in-plane conductivity of the MPL-coated GDL SGL 10BE (50% PTFE) was found to be higher than that of the counterpart SGL 10BC (25% PTFE) and this was explained by the presence of more conductive carbon particles in the MPL of SGL 10BE. Further, the conductivity of each GDL sample was measured in two perpendicular in-plane directions in order to investigate the in-plane anisotropy. The results show that the electrical conductivity of the GDL sample in one direction is different to that in the other direction by a factor of about two. The contact resistance, the main factor affecting the through-plane conductivity, of PTFE-treated GDLs shows a different trend to the corresponding in-plane conductivity, namely it increases as the PTFE loading increases. On the other hand, the contact resistance of the MPL-coated GDL SGL 10BE (50% PTFE) was found to be lower than that of the counterpart SGL 10BC (25% PTFE) and again this was explained by the presence of more conductive carbon particles in the MPL of SGL 10BE. Also, it was noted that the MPL coating appears to have a positive effect in reducing the contact resistance between the GDL and the bipolar plate. This is most likely due to the compressibility of the MPL layers that allows them to fill in the 'gaps' that exist in the surface of the bipolar plates and therefore establishes a good contact between the latter plates and the GDLs. Finally, good curve fitting of the contact resistance as a function of the clamping pressure has been achieved.

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1. Introduction

In proton exchange membrane fuel cells (PEMFCs), one of the major functions of the gas diffusion layer (GDL) is to transport electrons from the catalyst layer to the bipolar (or flow-field) plate and vice versa. This implies that the GDL must have a good electrical conductivity.

The electrical conductivity in the through-plane (or transverse) direction is usually reported by most suppliers of GDLs [1]. However, the electrical conductivity in the in-plane direction is as important as the transverse conductivity and this is due to that fact that the surface of the GDL is not entirely in contact with

the bipolar plate—which hosts the flow channels, see Fig. 1. As such, the in-plane conductivity can compensate for poor through-plane conductivity underneath the flow channels and thereby the evaluation of the conductivity in both directions is of paramount importance.

As mentioned above, the through-plane electrical conductivity (or resistivity) is usually reported by the GDL manufacturers as a measure of how conductive (or resistive) the GDL material is to the flow of electrons, see for example [2]. This measurement is typically achieved by fitting a sample of known surface area between two highly conductive electrodes (e.g. gold-plated electrodes). The subsequent step is to measure the resistance of the GDL sample to the electrical current by measuring the potential difference across the sample. It should be noted that the GDL sample is typically compressed by applying a pressure of 10 bar, which is the usual value of the clamping pressure in real PEM fuel cells [3], in order to minimise the contact (or interfacial) resistance between the components of the fuel cell.

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Nomenclature

A	curve fitting coefficient ($\Omega \text{ m}^2$)
B	curve fitting coefficient
C	correction factor
R	electrical resistance (Ω)
t	thickness of GDL sample (m)

Greek symbols

ε	porosity of the porous medium
ρ	electrical resistivity ($\Omega \text{ m}$)
σ	electrical conductivity (S m^{-1})

Although the in-plane conductivity is larger than the through-plane conductivity by an order of magnitude [1,4], it has received much less attention in the open literature. This lack of awareness of the in-plane conductivity manifests itself as a common assumption adopted by many PEMFC modellers that the electrical conductivity is isotropic and therefore its value in the in-plane direction has the same value to that reported (or measured) in the through-plane direction.

Recently, Zhou and Liu [5] showed that neglecting the anisotropic nature of the GDL leads to an overestimation of the GDL resistance and this, in turn, gives rise to an overestimation of the ohmic losses caused by this layer. This is fully understandable if one recognises that the considerably larger in-plane conductivity outweighs the smaller transverse conductivity. Therefore, there has been a recent trend to measure the conductivity of the GDLs in both the transverse and in-plane directions, e.g. [4]. Now, we present some publications whose authors measured and/or investigated the electrical conductivity of the GDLs.

Mathias et al. [4] measured the electrical resistivity of GDLs in both the transverse and in-plane directions and showed, as stated previously, that the in-plane resistivity is smaller than the through-plane one by an order of magnitude. Also, they emphasised that the contact resistance between the GDL and the bipolar plate is a major source of the ohmic losses in the through-plane direction. Thus, there have been recent works that have quantified the contact resistance [3,6–9] and computationally investigated its effect on the performance of PEM fuel cells [5,7,10–14].

On the other hand, Williams et al. [15] measured the resistivity in the in-plane direction by employing a four-point method suggested by ASTM C611 standard [16]. However, this standard is basically devised to measure the electrical resistivity of carbon

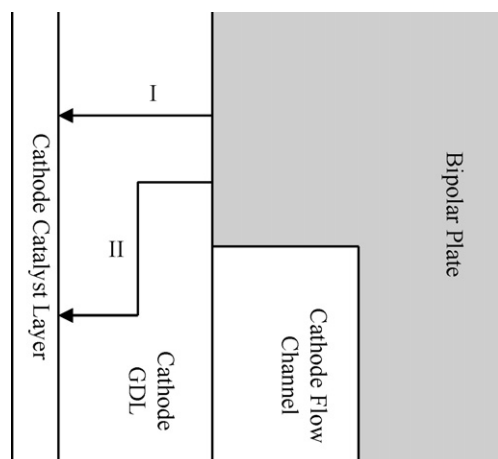


Fig. 1. A schematic diagram of possible paths for the electrical current through the GDL, I and II.

blocks, not thin carbon sheets. Thus, one may be dubious regarding the suitability of this standard to estimate the in-plane conductivity of the GDLs.

With the exception of the work published by Mathias et al. [4], it is notable that the anisotropy of the conductivity in the in-plane direction and also the effect of ‘enhancement’ processes (such as polytetrafluoroethylene (PTFE)-treatment and microporous layer (MPL)-coating) have not been investigated in the above works. Typically, the GDL is treated with PTFE to impart the hydrophobicity that it requires to expel excessive water present in the electrodes. Also, it is normal practice to deposit a thin layer (MPL), which consists of carbon and PTFE particles, in order to establish good contact between the GDL and other components of the PEM fuel cell, namely the catalyst layer and the bipolar plate.

Mathias et al. [4] found a discrepancy between conductivities measured in two different in-plane directions, although this observation was limited to one type of GDL: Toray TGP-H-060. Also, these authors investigated the effect of PTFE on the through-plane contact resistance. They found that this parameter increases when the GDL is treated with 3.5 wt.% PTFE. Again, this investigation was limited to the Toray GDL type mentioned above. Further, the authors did not investigate the effect of the PTFE-treatment on the in-plane conductivity. Moreover, the effect of the MPL coating on the conductivity of the GDL was not probed.

Hence, in this paper, the anisotropy of the electrical conductivity of GDLs and its sensitivity to PTFE-treatment and MPL coating have been investigated. Also, the through-plane contact resistance has been studied as a function of the clamping pressure.

2. Materials and methods

2.1. Materials

The electrical conductivity in both the through- and in-plane directions has been measured for five types of carbon substrates and two MPL-coated GDLs. The samples were supplied by SGL Technologies GmbH, Meitingen, Germany. The samples, and their parameters that are provided by the supplier, namely the PTFE loading and the areal weight, are listed in Tables 1 and 2, respectively. In addition, SEM imaging has been used to elucidate the surface morphology of the GDLs listed and to assist in interpreting some of the results. These images were obtained at the Leeds Electron Microscopy and Spectroscopy Centre, the University of Leeds, using a Philips XL30 ESEM (for micro-scale related images) and LEO 1530 Gemini FEGSEM (for nano-scale related images).

2.2. Setup

2.2.1. In-plane conductivity

The in-plane conductivity of the GDL samples was measured using the 4-probe method as described by Smits [1,17], who devel-

Table 1
Specifications of carbon substrates.

GDL	PTFE loading, wt.%	Areal weight, g m^{-2}
SGL 10AA	0	85.0
SGL 10BA	5	85.0
SGL 10CA	10	90.0
SGL 10DA	20	100.0
SGL 10EA	30	112.9

Table 2
Specifications of MPL-coated GDLs.

GDL	PTFE loading (MPL), wt.%	Areal weight, g m^{-2}
SGL 10BC	20–25	135
SGL 10BE	~50	139

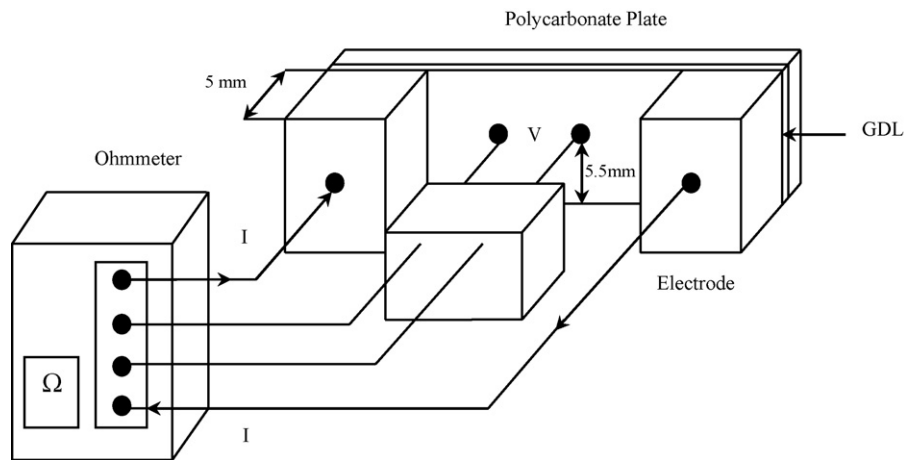


Fig. 2. A schematic diagram for the in-plane conductivity setup.

oped a formula that involves a geometry-dependent correction factor for thin sheets. Basically, the correction factor is a function of the dimensions of the sample and the space maintained between the probes. More details are presented in Section 2.3.

The tested GDL sample (11 mm width \times 53 mm long) was positioned on an insulating polycarbonate plate. The thickness of the GDLs varied from one GDL type to another and even from one sample to another. Further, there was a slight variation in the thickness of the GDL sample from one position to another. Therefore, the thickness of each GDL sample was measured by a micrometer at five equally spaced positions and the average value was then calculated. Copper electrodes (10 mm \times 10 mm \times 5 mm) were fixed at each end of the GDL sample with the assistance of G-clamps (not shown in the figure for the sake of clarity), see Fig. 2.

The electrical resistance was directly read from a BS401 Ohmmeter, which has a resolution of 0.01 m Ω . The equipment provides a current that passes through the sample via the copper electrodes and then measures the voltage, using two gold-plated probes, between two selected points located on the path of that electrical current. As such, the resistance is calculated using Ohm's Law. The distance between the voltage probes was kept constant during the course of the measurements. This was achieved by embedding the two probes in a rectangular plastic body. The spacing between the probes was 11.0 mm. Also, the probes were deliberately kept at a height 5.5 mm above the work surface (see Fig. 2), which is half the width of the tested GDL sample, in order for the measurements to be in compliance with Smits method. In addition, also in line with Smits method, the spacing between each probe and the electrode adjacent to it was adjusted so as to be the same as the spacing between the probes.

2.2.2. Through-plane conductivity

The through-plane conductivity of the tested GDLs was also evaluated by adopting the 4-probe method. A circular GDL sample (10 mm diameter) was placed between two 316-stainless steel discs (10 mm diameter \times 2 mm thick) that represent the bipolar plates. The sample and the discs were all sandwiched between two copper electrodes (10 mm diameter \times 10 mm thick). High-density nylon insulators were placed between the electrodes and the other metallic parts of the setup, namely the bolt and the load cell, as shown in Fig. 3. The bolt was used to compress the GDL in 10 progressively increasing increments. The compressive force was detected by a 10 kN load cell that was placed underneath the bottom insulator. The total resistance of the assembly was measured at each compressive pressure by a BS401 Ohmmeter, which has a resolution of 0.01 m Ω . The contact surfaces of the copper electrodes and the stainless steel bipolar plates were pol-

ished by 500-grade abrasive papers each time the GDL sample was replaced.

2.3. Data analysis

2.3.1. In-plane conductivity

As stated in Section 2.2, the correction factor in Smits method depends on the dimension of the GDLs and the spacing between the probes. Basically, it is a function of two ratios, namely the length of the GDL to its width (a/b), and the width of the GDL to the spacing between the probes (b/s), see Fig. 4. In this study, these ratios were found to be 5 and 1, respectively. This gives a value of 0.9994 (almost unity) to the correction factor [17]. The electrical resistivity, ρ , can be then calculated using the following formula [17]:

$$\rho = CtR, \quad (1)$$

where C is the correction factor, t is the thickness of the GDL, and R is the electrical resistance. The reciprocal of the electrical resistivity

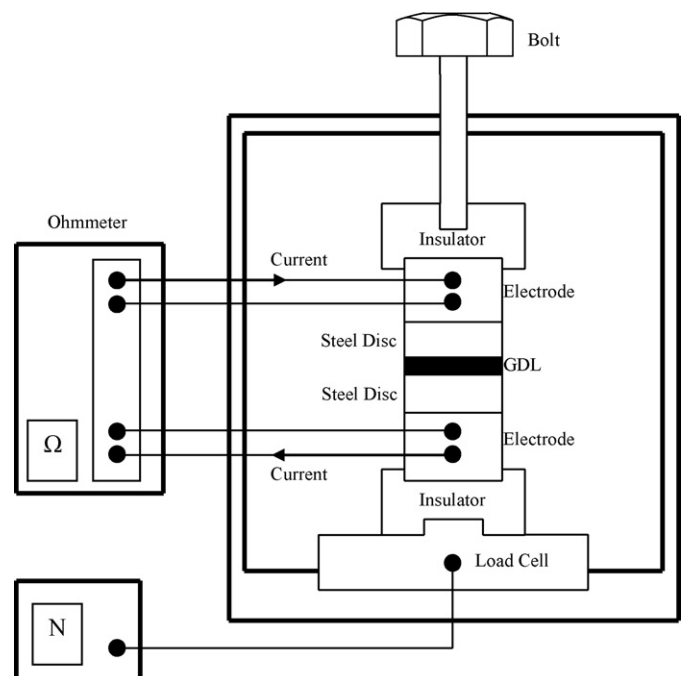


Fig. 3. A schematic diagram for the through-plane conductivity setup.

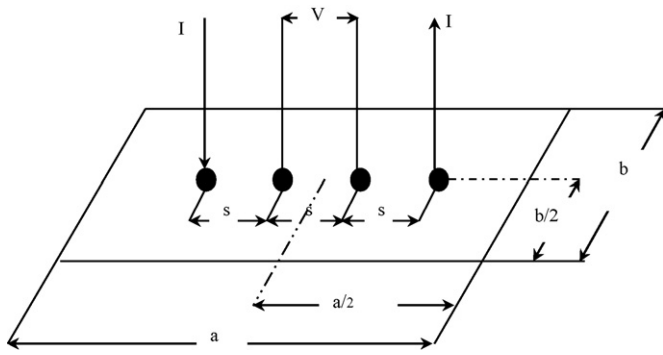


Fig. 4. Arrangement of the four probes on a rectangular sample according to Smits method.

gives the electrical conductivity σ :

$$\sigma = \frac{1}{\rho}. \quad (2)$$

The in-plane conductivity was measured for six samples taken from the same GDL sheets, three of which were cut off in a direction perpendicular to that of the first three samples. This was made in order to investigate the anisotropy of the GDL. The electrical conductivity of each GDL sample was measured eight times, four of which were when the direction of the current was reversed. The individual values of the resistance were then averaged.

2.3.2. Through-plane conductivity

The contact resistance between the GDL and the bipolar plate is one of the major sources of the ohmic losses experienced by PEM fuel cells. The bulk resistance of the GDL is considerably smaller in comparison to the contact resistance [4]. Therefore, one can understand why the objective of most of the previous publications in this regard was to investigate and/or quantify the contact resistance, see for example [7,11–14]. It should be noted that such investigations allow the researcher(s) to equally evaluate the material from which the GDLs and the bipolar plates are made. As a consequence, the design of the currently used material may be modified or new materials may be suggested in order to minimise the contact resistance between the two major components of PEM fuel cells, namely the GDL and the bipolar plate. Therefore, in the present work, studies on the electrical conductivity in the through-plane direction will be limited to quantifying the contact resistance between the various types of GDLs and a bipolar plate, which is made from one of the most commonly used materials, 316 stainless steel. In order to quantify the contact resistance, the two arrangements shown in Fig. 5 were considered.

In arrangement (a), the total resistance R_1 is a combination of the following resistances:

$$R_1 = 2R_{Cu} + 2R_{Cu-st} + 2R_{st} + 2R_{st-GDL} + R_{GDL}, \quad (3)$$

where R_{Cu} , R_{st} , and R_{GDL} are the bulk resistances of the copper electrode, the stainless steel bipolar plate, and the GDL, respectively. R_{Cu-st} is the contact resistance between the copper electrode and the bipolar plate and R_{st-GDL} is the contact resistance between the bipolar plate and the GDL. Likewise, the total resistance R_2 in arrangement (b) can be expressed as follows:

$$R_2 = 2R_{Cu} + 2R_{Cu-st} + R_{st}. \quad (4)$$

Subtraction of Eq. (4) from Eq. (3) results in a formula to calculate the contact resistance R_{st-GDL} :

$$R_{st-GDL} = \frac{1}{2}(R_1 - R_2 - R_{st} - R_{GDL}). \quad (5)$$

Both total resistances R_1 and R_2 are measured by the setup shown in Fig. 3 or by this setup without one of the stainless steel discs and

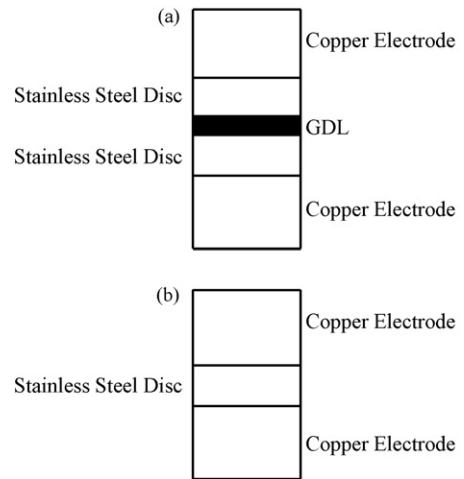


Fig. 5. Arrangements (a) and (b) that have been considered to estimate the contact resistance between the GDL and the bipolar plate.

without the GDL, respectively. The bulk resistance of the bipolar plate R_{st} can be obtained if the resistivity of 316-stainless steel is multiplied by the thickness of the bipolar plate, viz. 2.0 mm. The resistivity of 316-stainless steel is $7.3 \times 10^{-7} \Omega m$ [18], and therefore the bulk resistance of the bipolar plate is about $0.015 m\Omega cm^2$.

The bulk resistance of the GDL can be obtained in the same manner. However, the GDL is a highly porous material and hence it is not a purely single material. Basically, air and carbon fibre constitute the material of the GDL. As such, the volume fraction weighted harmonic mean of the resistivity of air and the carbon fibre was used in order to estimate the bulk resistivity of the GDL, ρ_{GDL} [3]:

$$\rho_{GDL} = \frac{1}{(\varepsilon/\rho_{air}) + ((1 - \varepsilon)/\rho_f)}, \quad (6)$$

where ε is the porosity of the GDL, and ρ_{air} and ρ_f are the resistivities of air and carbon fibre, respectively. The first term in the denominator in Eq. (6) approaches zero as the resistivity of air is extremely high. As a consequence, Eq. (6) approximates to:

$$\rho_{GDL} = \frac{\rho_f}{1 - \varepsilon}. \quad (7)$$

The resistivity of carbon fibre used by [3] was calculated to be $4.02 \times 10^{-5} \Omega m$. The porosity of the GDL can be calculated using the following formula [1,19]:

$$\varepsilon = 1 - \left(\frac{\text{Areal weight (g m}^{-2}\text{)}}{\text{Thickness (mm)} \times 1.8 \text{ (g cm}^{-3}\text{)}} \right). \quad (8)$$

The areal weights of the tested GDLs were listed in Tables 1 and 2. The thickness of each GDL was measured five times and the average value was then calculated. Substituting the values of ε and ρ_f into Eq. (7) gives the resistivity of the GDL which can be converted to bulk resistance if multiplied by the thickness of the GDL. It should be noted that the bulk resistance of the tested GDLs were found to be small (the order of $1 m\Omega cm^2$) and thereby the change in that resistance when compressing the GDL was ignored.

As mentioned in Section 2.2, the GDL sample was examined under 10 different gradually increasing compressive pressures. The total resistance at each compressive pressure was measured eight times, four of which were when the direction of the current was reversed. The individual readings were then averaged. These measurements were taken for five separate samples of each of the GDL materials investigated. The resulting values for the contact resistance R_{st-GDL} were then averaged and the 95% confidence interval was calculated around the mean value.

3. Results and discussion

3.1. In-plane conductivity

Fig. 6 presents the in-plane electrical conductivity of the tested PTFE-treated GDLs as a function of the PTFE content in the two principal directions. As mentioned in Section 2.3, three samples of each GDL were examined in each in-plane direction. It might be expected that the conductivity would decrease with an increase in the PTFE loading; however it is almost constant in both directions. This is most likely due to the fact that the GDL is basically constituted of electrically conductive carbon fibres which remain structurally unaltered when treating the GDL with PTFE. As such, the bulk conductivity is not expected to change after the PTFE-treatment. Fig. 7 shows 150 \times magnification SEM images of the tested PTFE-treated GDLs. Apparently, treating the bare GDL, SGL 10AA, with various amounts of PTFE does not change the structure of the GDL. It is of particular importance not to confuse the resin

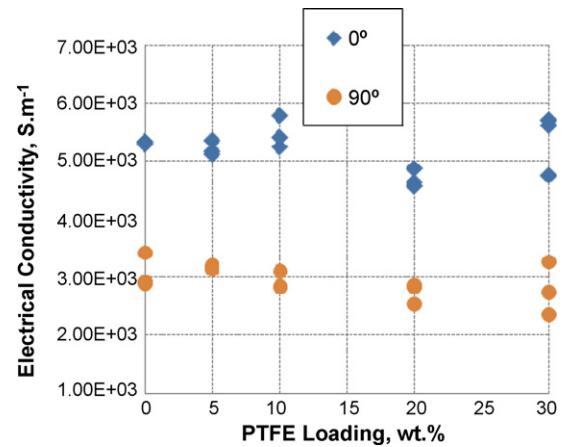


Fig. 6. In-plane conductivity of the tested PTFE-treated GDLs. The two angles in the legend label indicate the orientation of the fibres in the GDL sample.

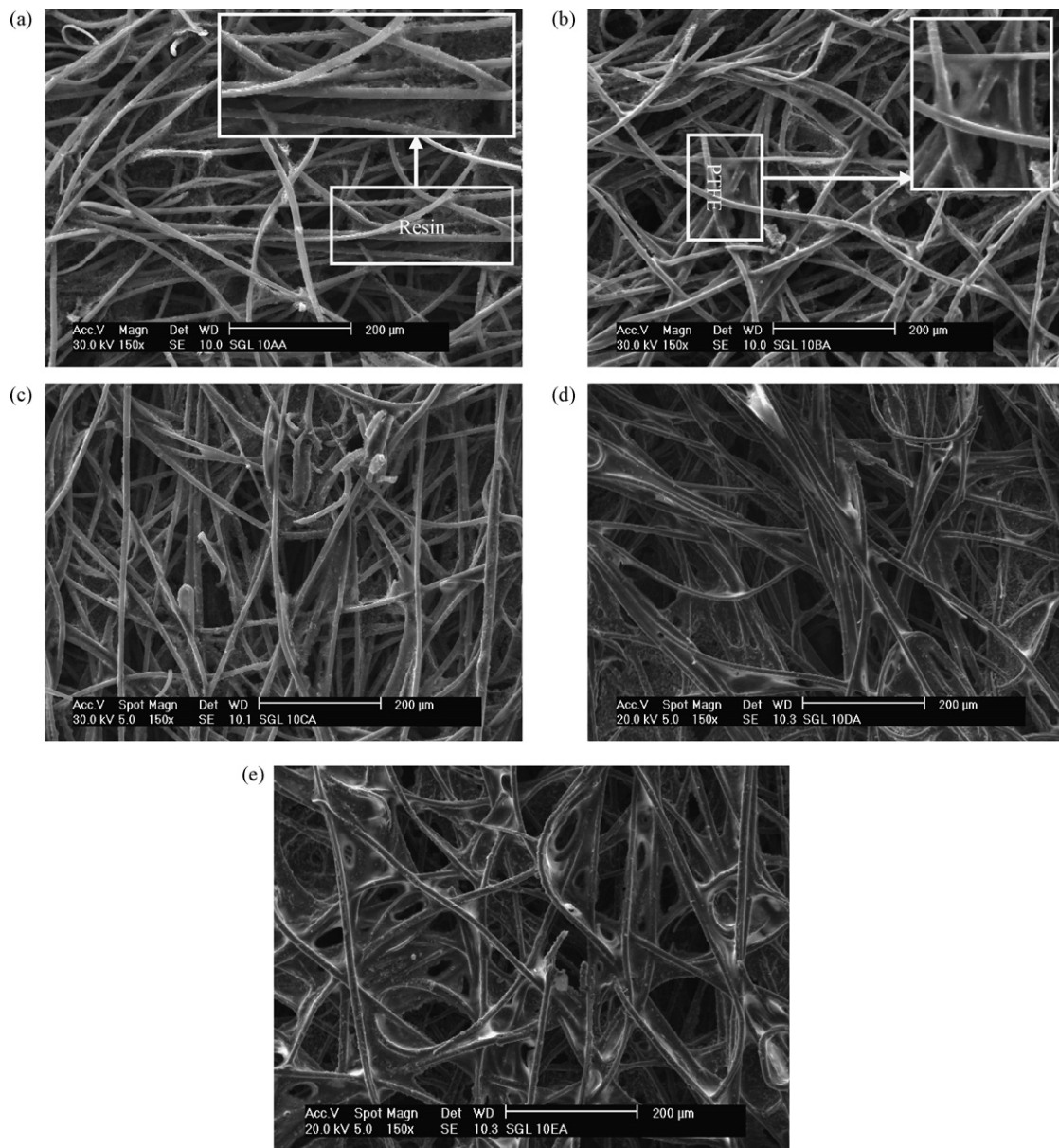


Fig. 7. SEM Micrographs for (a) SGL 10AA, (b) SGL 10BA, (c) SGL 10CA, (d) SGL 10DA, and (e) SGL 10EA GDLs. The images at upper left corners in (a) and (b) show magnified pictures of resin and PTFE, respectively.

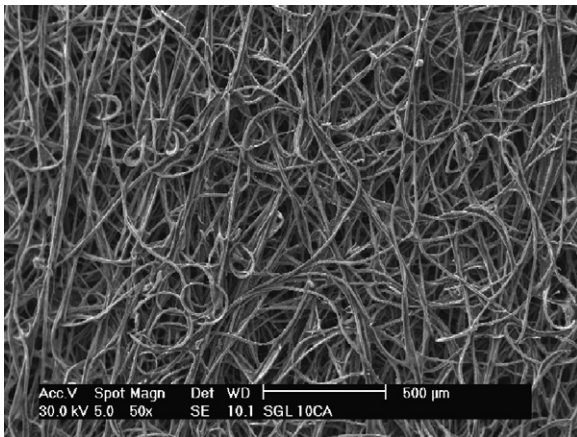


Fig. 8. 50× magnification SEM image for SGL 10CA.

used to bind carbon fibres with the PTFE clusters. Typically, the latter clusters (shown in Fig. 7b) have a smooth surface in comparison with the resin material (see Fig. 7a, the SEM micrograph of SGL 10AA, which only contains resin). Also, it is of interest to note that careful examination of the SEM images in Fig. 7 shows that a considerable amount of PTFE material tends to cluster in the gaps between and at the intersections of carbon fibres. Where the fibre surfaces appear to be of the same brightness to that of the previously mentioned PTFE clusters, this is an indication that these fibre surfaces are coated with PTFE. On the other hand, the fibre surfaces that remain of higher brightness appear to remain without this coating of PTFE. Basically, one can be more certain regarding the coating of the fibres with PTFE if the energy-dispersive X-ray spectroscopy (EDX) is utilised to chemically characterise the fibre surface. The PTFE-coated surface must give a signal that is characteristic of the fluorine, F.

Further, Fig. 6 shows that the conductivities in two perpendicular in-plane directions differ by a factor of about two. This is most likely due to the fact that the fibres of the GDLs are preferentially more aligned in one of the in-plane directions. Fig. 8 shows a 50× magnification SEM micrograph of SGL 10CA where the preferential alignment of most carbon fibres in one in-plane direction is reasonably clear. Such a trend can be observed in all other PTFE-treated GDLs. In general, one can observe that the conductivities in the first direction (designated as 0°) and the second direction (designated as 90°) are scattered around 5000 and 3000 S m⁻¹, respectively. These results clearly show that the assumption that the conductivity in the in-plane direction is isotropic is not valid. This might be of interest to modellers of PEMFCs, of whom many have assumed that the electrical conductivity of the GDL is isotropic. The above results clearly show that the in-plane conductivity of the tested GDLs is not only different to that in the through-plane direction (typically by an order of magnitude [4]), but also significantly different from one in-plane direction to another. Capturing such anisotropic nature of the electrical conductivity of the GDLs will likely enhance the predictive ability of the models built.

As mentioned in Section 1, most publications in the literature have focused on estimating the through-plane (or transverse) conductivity. Therefore, there is virtually no reported value for the in-plane conductivity of the tested GDLs to compare with. However, Williams et al. [15] reported that the conductivity of SGL 10BA in the in-plane direction is about 4800 S m⁻¹, which is comparable to that measured in the 0° direction in the present study for the same GDL (about 5200 S m⁻¹). It should be noted that Williams et al. [15] did not mention the orientation of the fibres constituting the GDL when measuring the in-plane conductivity.

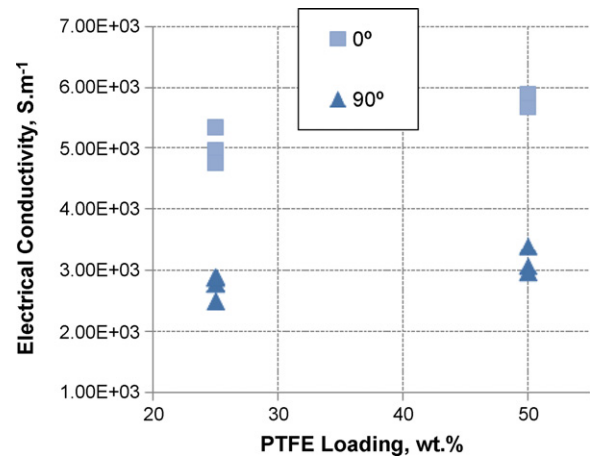


Fig. 9. In-plane conductivity of the tested MPL-coated GDLs. The two angles in the legend label indicate the orientation of the fibres in the GDL sample. The PTFE content in the MPL of SGL 10BC does not necessarily have an indeliberately selected value of 25 wt.% (it might be between 20 and 25 wt.%).

Furthermore, the tested MPL-coated GDLs show an unexpected trend as the in-plane conductivity of SGL 10BE is higher than that of SGL 10BC despite the fact that the amount of electrically insulating PTFE in the MPL of the latter GDL (20–25 wt.%) is significantly lower than that in the former one (50 wt.%), see Fig. 9. This may be explained by referring to the SEM images of the respective GDLs (see Fig. 10) which suggest that more (presumably) electrically conductive carbon particles are present in the MPL of SGL 10BE when compared with that of SGL 10BC. It should be noted that the areal weights of the above two MPL-coated GDLs are comparable, see Table 2. Therefore one would not expect the carbon loading of SGL 10BE to be significantly larger than that of SGL 10BC. However, it should be mentioned that the carbon substrate used in the above GDLs, viz. SGL 10BA, may differ from batch to batch. The authors measured the thicknesses of SGL 10BC and SGL 10BE to be about 420 and 390 μm, respectively. Hence, it may be inferred that the presumably lower mass of carbon fibres in SGL 10BE (compared to that of SGL 10BC) is outweighed by an apparently higher quantity of carbon particles which, presumably, enhance the electrical conductivity. One more possible reason for having more carbon particles on the surface of the SGL 10BE compared to that of SGL 10BC is that more MPL constituents (i.e. PTFE and carbon particles) may have penetrated to the carbon substrate of the latter GDL while performing the coating. However, this merits a future experimental investigation that elucidates the microstructure of the cross-sectional area of the GDL—probably via SEM images of cross-sectional areas of the MPL-coated GDLs.

As mentioned in Section 2.3, the Smits method involves a factor that corrects for the dimensions of thin sheets. Ideally, the electrical conductivities of GDL samples having different dimensions should all have the same value, as the electrical conductivity is an intrinsic property of the material. The in-plane electrical conductivity of a sample (namely SGL 10AA, in 90° orientation) of different dimensions to those adopted in the present study was measured in order to evaluate the suitability of the Smits method for GDL materials. The width of the sample (*b*) has been made to be 13.8 mm (rather than 11.0 mm) so that the ratio of the width of the sample to the spacing between the probes (*b/s*) became 1.25 (see Fig. 4). As such, the correction factor *C* is 1.2248 [17]. This gives an electrical conductivity of about 3394 S m⁻¹ if Eqs. (1) and (2) are employed. Such a value lies between the limits reported for the in-plane electrical conductivity of SGL 10AA in 90° orientation, see Fig. 6. Therefore, it can be concluded that the Smits method appears to be suitable to estimate the electrical conductivity of GDLs.

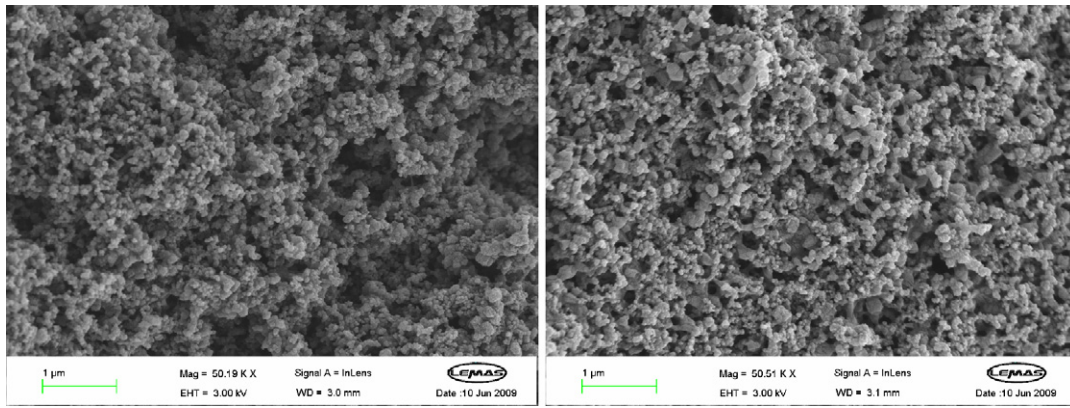


Fig. 10. SEM images at about 50,000 \times magnification of the MPLs in SGL 10BC (left) and SGL 10BE (right).

3.2. Through-plane conductivity

Fig. 11 shows the contact resistance between a bipolar plate and five GDLs with various PTFE loadings as a function of clamping pressure. The error bar estimates the 95% confidence interval calculated for five samples. In contrast to the behaviour observed in the in-plane direction, one can observe that a clear relationship exists between the PTFE loading and the contact resistance of the GDL in the through-plane direction in that the latter increases with increasing PTFE loading. However, it can be seen that there are insignificant differences in contact resistances between SGL 10DA and SGL 10 EA (i.e. smaller than the respective error bars). It might be possible that both of them have a comparable 'real' amount of PTFE. However, such an argument would require further tests from which the actual amount of PTFE can be estimated, such as hydrophobicity-related tests. As expected, contact resistance decreases with increasing clamping pressure as voids and air-gaps are closed up.

Fig. 12a shows the contact resistance between the two types of MPL-coated GDLs and the bipolar plate. Once again, the error bars estimate the 95% confidence interval calculated for the five samples. Although the PTFE loading in the MPL of SGL 10BE is higher than that in SGL 10BC, the contact resistance of the former GDL is lower than that in the latter. This may be explained by the fact, as mentioned previously, that more (presumably) electrically conductive carbon particles are present in the MPL of SGL 10BE when compared with that of SGL 10BC, as shown in Fig. 10. In addition, it should be noted

that the PTFE clusters shown in Fig. 10 have smooth surfaces. Thus, the surface in the MPL-coated side of SGL 10BE (50 wt.% PTFE) may have less roughness than that of SGL 10BC (20–25 wt.% PTFE).

In addition, coating of the carbon substrates with MPLs appears to increase the through-plane conductivity and this is most likely due to the compressibility of the MPL layers and, in turn, their ability to fill in the gaps present in the surface of the bipolar plates. Fig. 12b clearly shows that the contact resistance of the carbon substrate SGL 10BA decreases by about 15% and 40% at 10 bar (the pressure at which the fuel cell is normally clamped) if it is coated

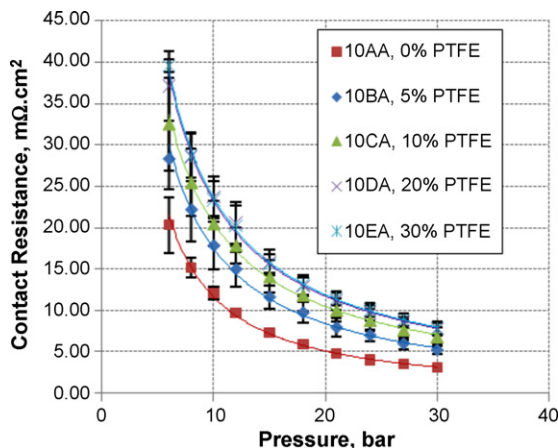


Fig. 11. Contact resistance as a function of clamping pressures for the tested PTFE-treated GDLs.

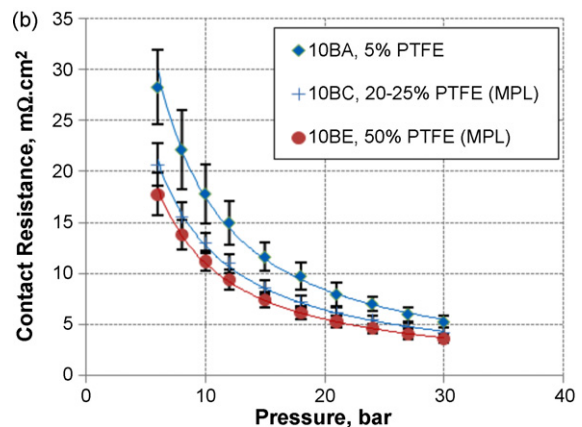
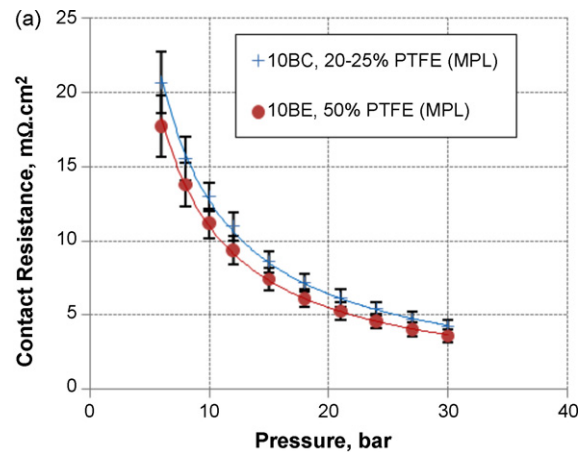


Fig. 12. (a) Contact resistance as a function of clamping pressure for the tested MPL-coated GDLs and (b) the effect of MPL coating on the contact resistance of carbon substrate SGL 10BA.

Table 3
Values of the coefficients A and B for the fitted curves.

GDL	A, $\Omega \text{ cm}^2 \text{ bar}$	B
SGL 10AA	182.03	1.194
SGL 10BA	198.73	1.056
SGL 10CA	191.71	0.970
SGL 10DA	226.65	0.989
SGL 10EA	228.51	0.987
SGL 10BC	123.59	0.987
SGL 10BE	109.91	1.000

with MPL layers to form SGL 10BC and SGL 10BE GDLs, respectively. It is possible that the carbon particles present in the SGL 10BC MPL, and present at higher concentration in SGL 10BE MPL, explain these results.

In spite of the fact that the MPL is typically in contact with the catalyst layer, not with the bipolar plate, there has been a recent trend to deposit MPLs on both sides of the GDL [20,21]. As such, quantifying the contact resistance between the MPL and the bipolar plate appears to be justifiably relevant.

The literature has shown a number of investigations which have reported the contact resistance of 316-stainless steel bipolar plates, see for example [22,23]. However, one should note that the measured contact resistance varies from one report to another. For example, Davies et al. [22] measured the contact resistance of 316-stainless bipolar plates to be about $75 \text{ m}\Omega \text{ cm}^2$ at 10 bar whereas Makkus et al. [23] reported a contact resistance of about $29 \text{ m}\Omega \text{ cm}^2$ for the same material and at the same compaction pressure. These discrepancies are due to using different GDLs and/or adopting different experimental procedures. As an example, the GDL used by Davies et al. [22] was Carbel CL while an E-tek GDL was employed by Makkus et al. [23]. As such, it would be misleading to compare the reported contact resistances of the above two GDLs, whose PTFE contents are also unreported, to that of the GDLs tested in the present study.

The curves of the contact resistance as a function of the clamping pressure in Figs. 11 and 12 present a decay-like trend. Therefore, the experimental data were curve-fitted empirically as follows:

$$R = AP^{-B}, \quad (9)$$

where R is the resistance, P is the pressure, and A and B are coefficients that are determined from numerically fitting with the experimental data. The experimental data shows good agreement with the resulting fitting curves in all cases, see Figs. 11 and 12. The curve fitting values of A and B are given in Table 3.

4. Conclusions

The electrical conductivities in both the through- and in-plane directions have been measured for a number of untreated, PTFE-treated, and MPL-coated GDLs. The in-plane conductivity was measured using the Smits method, which involves a geometry-dependent correction factor. The results show that the conductivity of PTFE-treated GDLs in the in-plane direction is almost constant, regardless of the PTFE loading. This can be attributed to the fact that the structure of the carbon fibres constituting the carbon substrates remains unaltered when adding the PTFE particles and therefore the bulk conductivities of the PTFE-treated GDLs are not expected to change. On the other hand, the in-plane conductivity of the MPL-coated GDLs was found to be higher in the SGL 10BE (50% PTFE loading) compared to that of the SGL 10BC (25% PTFE) and this can be explained by the presence of more conductive carbon particles in the MPL of SGL 10BE.

Significant anisotropy of the electrical conductivity in the in-plane direction was found to be present. The conductivity in one direction can differ from that in the perpendicular direction by a factor of about two, namely 5000 and 3000 S m^{-1} . This is most likely due to the fact that the fibres of the GDLs are preferentially aligned in one in-plane direction. Therefore, the assumption that the conductivity in the in-plane direction is isotropic appears not to be valid.

In contrast to the in-plane conductivity of the PTFE-treated GDLs, the contact resistance, which is the major factor affecting the conductivity in the through-plane direction, appears to have a clear relationship with the PTFE loading—it was found to increase with an increase in the amount of PTFE owing to its electrically insulating effect. The bulk through-plane resistance was found to be very small compared to the respective contact resistance and therefore the change in that resistance when compressing the GDL has been neglected in the calculations. On the other hand, the contact resistance of the MPL-coated GDL SGL 10BE (50% PTFE) was found to be lower than that of the counterpart SGL 10BC (25% PTFE). Again, this result was explained by the presence of more conductive carbon particles in the MPL of SGL 10BE. Also, it appears that the MPL coating has a positive effect in reducing the contact resistance of carbon substrates. This is mostly due to the compressible nature of the MPL layers that allows them to fill in the gaps exist in the surface of the bipolar plate and accordingly enhance the contact. Finally, the experimental contact resistance data for the GDLs as a function of the clamping pressure shows a decay-like trend. This experimental data has been empirically curve-fitted with good agreement being achieved between the experimental and fitted curves.

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