

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/228408496>

Microstructure Analysis and Calculation of Thermal Conductivity of Gas Diffusion Layers of PEM Fuel Cells

Article · August 2009

CITATIONS

0

READS

59

2 authors:



[Andreas Pfrang](#)

European Commission

40 PUBLICATIONS 353 CITATIONS

[SEE PROFILE](#)



[Damien Veyret](#)

Polytech Marseille

49 PUBLICATIONS 754 CITATIONS

[SEE PROFILE](#)

Microstructure Analysis and Calculation of Thermal Conductivity of Gas Diffusion Layers of PEM Fuel Cells

Andreas Pfrang, Damien Veyret

European Commission, DG Joint Research Centre, Institute for Energy
Cleaner Energy Unit
Post Office Box 2
NL-1755 ZG Petten / The Netherlands
Tel.: +31-224-56-5047
Fax: +31-224-56-5625
andreas.pfrang@ec.europa.eu

Abstract

The effective thermal conductivity of gas diffusion layers (GDL) is an important parameter for the analysis of polymer electrolyte membrane (PEM) fuel cells as thermal conductivity strongly influences fuel cell performance. The accuracy of modeling heat transfer – and therefore also performance – in a PEM fuel cell relies on the accurate estimation of effective thermal conductivity.

Commercially available gas diffusion layers were investigated by 3D x-ray computed tomography (CT). Based on the 3D structure reconstructed from tomography data, the macroscopic effective thermal conductivity of the gas diffusion layers was calculated by solving the energy equation considering a pure thermal conduction problem.

Introduction

In polymer electrolyte membrane fuel cells (PEMFC) gas diffusion layers (GDL) – in combination with bipolar plates – are usually employed to distribute the reactants over the electrode surfaces. The GDL has not only the function to provide gas access from the flow field channels of the bipolar plate to the catalyst layer, but also to allow the removal of the product water, to mechanically stabilize the membrane-electrode assembly without blocking the flow field channels, to provide electronic and finally also thermal conductivity between catalyst layer and bipolar plate [1]. In PEMFC, usually porous carbon fiber papers or cloths infiltrated with polytetrafluoro ethylene (PTFE) are used as gas diffusion layers. Accurate information about the typically anisotropic thermal conductivity of the GDL is crucial for the optimization of heat and water management as the GDL has a big influence on temperature distribution and heat transfer in the fuel cell.

The thermal conductivity of a gas diffusion layer can either be measured directly [2-4] or calculated based on structural 3D data of the GDL [5, 6]. This structural data can either be derived from a structural model or measured experimentally. One of the suitable techniques for three dimensional volume imaging of porous structures is x-ray computed tomography (CT). It is non-destructive and can reach a resolution well below 1 μm which is sufficient for the imaging of carbon fibers with typical diameters between 5 and 10 μm . CT has been used for the investigation of PEMFC membranes [7] as well as for the investigation of wetting behavior of gas diffusion layers [8, 9]. In this investigation, the 3D structure of gas diffusion layers was determined by x-ray computed tomography and based on this structural data, the thermal conductivity was calculated.

1. Experimental

Two different carbon fiber based gas diffusion layers were chosen as samples: the carbon cloth EC-CC1-060T (thickness 0.33 mm) and the Toray carbon paper EC-TP1-060T [10] (thickness 0.19 mm). Both samples are available from ElectroChem, Woburn, USA and were treated with 30 wt% of PTFE by the manufacturer.

A strip with a width of about 3 mm was cut from each gas diffusion layer. These strips were investigated by a nanotom[®] x-ray computed tomography system (GE Sensing & Inspection Technologies, phoenix x-ray, Wunstorf, Germany). This system includes a 180 kV x-ray tube and a 2 D x-ray detector with 2300 x 2300 pixels. The chosen voxel size was 0.7 μm for the carbon paper EC-TP1-060T and 1.4 μm for the carbon cloth EC-CC1-060T. In plane-view images, the lateral coordinates are denoted as x (direction bottom-top) and y (left-right). The direction perpendicular to the image plane – and accordingly perpendicular to the gas diffusion layer – is denoted as z.

The computation of the effective thermal conductivity of fibrous materials requires the resolution of the steady, purely diffusive, three-dimensional heat transfer equation; convection and radiation transport, as well as thermal contact resistance and phase changes are not taken into account. In the case of large three-dimensional geometries, generated randomly or based on large data sets from digital imaging (like computer tomography), partial differential equation solvers are not efficient. The description of these complex geometries combined with the large difference in thermal conductivity of the matrix and fibers demand a very fine mesh, and so large amount of memory and CPU time. Wiegmann & al. [11] use a different approach: the energy equation is solved by harmonic averaging and explicitly introducing jumps across the material interfaces as additional variables. These extra variables can be determined through the continuity of

heat fluxes at the interfaces. FFT and BiCGStab methods are then used to solve the Schur-complement formulation for the new variables. This method is implemented in an integrated simulation environment named GeoDict (Fraunhofer ITWM, Kaiserslautern, Germany, [12]). GeoDict was used in the current study to compute the effective thermal conductivity in x, y and z direction.

All solid voxels were assumed to have a thermal conductivity of $120 \text{ W m}^{-1} \text{ K}^{-1}$, which is the value of the thermal conductivity of carbon fibers [3], i.e. the thermal conductivity of PTFE of ca. $0.23 \text{ W m}^{-1} \text{ K}^{-1}$ [3] or any binder material was not taken into account as the PTFE or binder could not be distinguished from the carbon fibers in the CT data. The remaining voxels were assumed to be filled with air (thermal conductivity: $0.024 \text{ W m}^{-1} \text{ K}^{-1}$).

2. Results

Fig. 1 shows reconstructions of the CT data of the two investigated gas diffusion layers. It is obvious that the resolution in both cases is sufficient to resolve the carbon fibers.

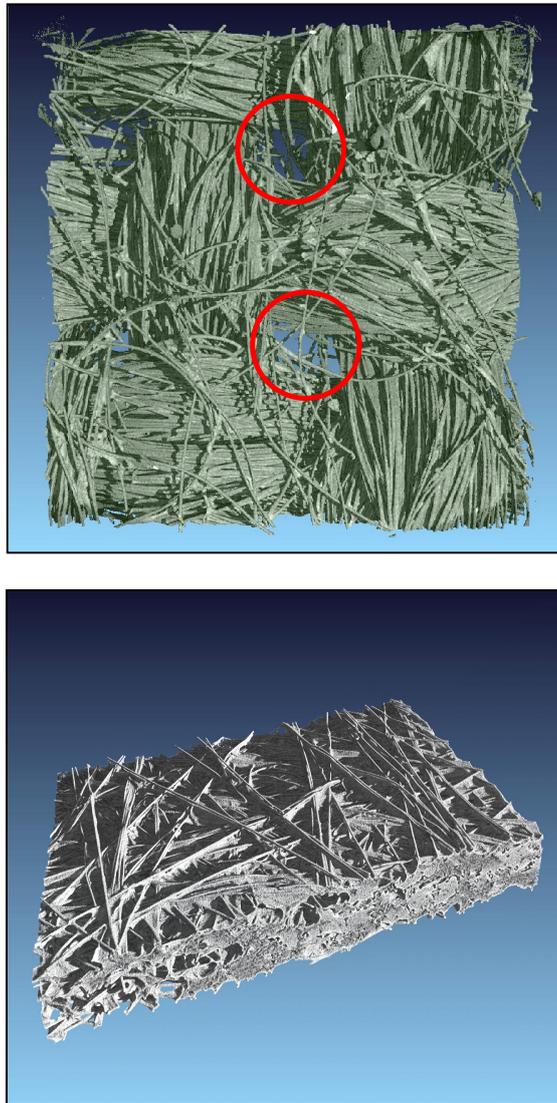


Fig. 1: X-ray computed tomography reconstruction of gas diffusion layers, top: carbon cloth EC-CC1-060T, imaged volume $1400 \mu\text{m} \times 1400 \mu\text{m} \times \text{ca. } 400 \mu\text{m}$, bottom: carbon paper EC-TP1-060T, imaged volume $1606 \mu\text{m} \times 1218 \mu\text{m} \times \text{ca. } 200 \mu\text{m}$.

The carbon cloth (top) contains mainly curved fibers while the fibers in the carbon paper (bottom) are predominantly coplanar and straight. Additionally, in both images the PTFE can be seen. As expected, PTFE forms frequently wing-like structures where two carbon fibers cross or run close to each other.

Furthermore it is obvious that the carbon cloth shows certain regions where almost no carbon fibers are present, i.e. where no threads of carbon fibers run (e.g. marked by the red circles in the top image of Fig. 1). This is not the case for the carbon paper.

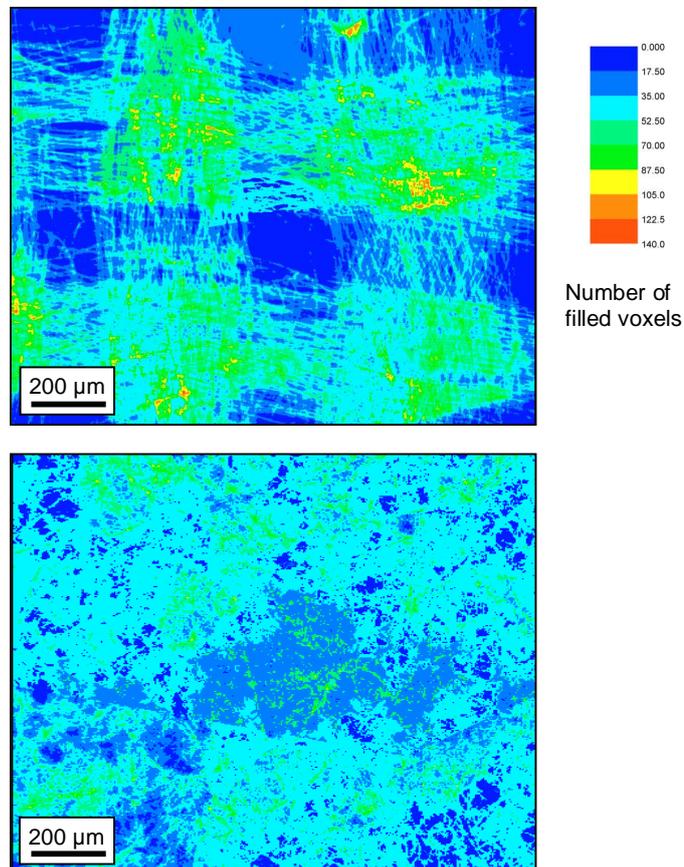


Fig. 2: Number of voxels filled with material for each lateral x-y-coordinate, top: carbon cloth EC-CC1-060T, bottom: carbon paper EC-TP1-060T.

Fig. 2 illustrates this observation more clearly as it shows for every lateral x-y-coordinate how many voxels are filled in the z-direction, i.e. for the whole thickness of the GDL. The same color scale is used for both samples. The number of filled voxels varies in a much wider range for the carbon cloth (top image).

Nine volumes were selected from the data of the carbon cloth for the calculation of the thermal conductivity as shown in Fig. 3. Each of these volumes included the whole thickness of the cloth. It is clear that the porosity as well as the preferred orientation of the fibers are different for the different volumes.

Table 1 shows the respective values for the thermal conductivity for x, y and z-direction together with the experimentally determined filling factors of each of the selected volumes. The filling factor is defined as the ratio of filled volume to total volume.

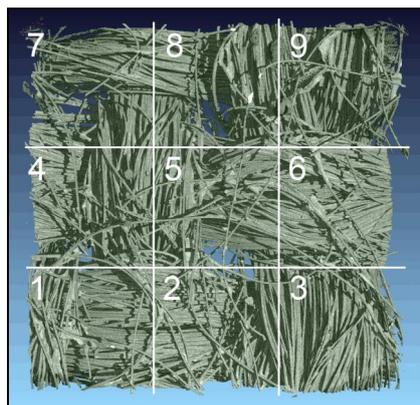


Fig. 3: Volumes selected for the calculation of thermal conductivity for the carbon cloth EC-CC1-060T.

Volume No.	Thermal conductivity / $W m^{-1} K^{-1}$			Filling factor
	X	Y	Z	
1	3.76	5.07	0.53	0.19
2	2.30	3.28	0.42	0.16
3	3.44	3.44	0.52	0.17
4	4.11	3.49	0.88	0.15
5	3.45	2.24	1.01	0.16
6	5.22	6.71	0.73	0.19
7	1.85	1.24	0.24	0.11
8	2.15	2.87	0.28	0.13
9	1.87	1.29	0.23	0.13
Average	3.13	3.29	0.54	0.16
Standard deviation	1.16	1.75	0.28	0.03

Table 1: Thermal conductivities calculated for the different parts of carbon cloth EC-CC1-060T (compare Fig. 3) together with the corresponding filling factors, voxel size $1.4 \mu m$.

Voxel size		Thermal conductivity / $W m^{-1} K^{-1}$		
		X	Y	Z
0.7 μm	Average	13.17	2.08	13.19
	Standard deviation	3.24	1.30	3.03
1.4 μm	Average	11.81	1.79	11.65
	Standard deviation	3.19	1.01	2.96
2.1 μm	Average	10.69	1.68	10.38
	Standard deviation	3.31	0.93	3.04
2.8 μm	Average	9.63	1.49	9.27
	Standard deviation	3.58	0.86	3.27

Table 2: Thermal conductivities calculated for the carbon paper EC-TP1-060T for different resolutions of the CT data. The standard deviations for each are calculated from the values originating from the same 25 different volumes of the carbon paper.

The thermal conductivities in z-direction are lower than the thermal conductivities in x- or y-direction. The values for the thermal conductivity in x- or y-direction vary between 1.24 and 6.71 W m⁻¹ K⁻¹ whereas the thermal conductivity in z-direction varies between 0.23 and 1.01 W m⁻¹ K⁻¹. The average thermal conductivities are 3.13, 3.29 and 0.54 W m⁻¹ K⁻¹ for x-, y- and z-direction, respectively.

Table 2 shows the respective values for the thermal conductivity for x, y and z-direction for the carbon paper EC-TP1-060T for different virtual resolutions of the CT data based on the measurements for 0.7 micron. The filling factor for this voxel size of 0.7 μm was 0.29. For the other – virtual – resolutions, the intensities for the voxels were simulated by adding up the intensities of 2³, 3³ or 4³ voxels, respectively. Then, the intensity threshold – above which a voxel is assigned to solid material – was chosen according to the criterion that the same filling factor of 0.29 as for the smallest voxel size must be achieved. Based on these structures, the thermal conductivities were calculated. Again, the thermal conductivities in z-direction are lower than the thermal conductivities in x- or y-direction. The thermal conductivities decrease with increasing voxel size.

3. Discussion

Carbon fibers are highly conductive, but in common gas diffusion layers they are mostly mainly oriented perpendicular to the GDL i.e. also perpendicular to the dominant direction of heat flow. Furthermore, it can be expected that the thermal conductivity parallel to the plane will be dominated by the conductivity of the fibers while the through plane conductivity will depend strongly on the contact between the fibers. Overall, this results in a low effective through plane thermal conductivity.

This is in agreement with the calculated results: The average in plane thermal conductivity is for both samples – the carbon cloth and the carbon paper – by about a factor 6 to 7 larger than the average through plane thermal conductivity. The average in plane thermal conductivities in x- and y-direction agree well for both samples within the expected range of statistical variation.

As expected, a clear dependence of thermal conductivity on the filling factor is observed in both samples: a higher filling factor leads to higher thermal conductivities as the thermal conductivity of carbon fibers is much higher than the thermal conductivity of air. This leads for example to relatively low thermal conductivities for volume 7 of the carbon cloth (compare Fig. 3 and Table 1).

Furthermore, also a dependence of thermal conductivity on the local orientation of the fibers was observed, e.g. in volume 5 of the carbon cloth (Fig. 3) the fibers are preferentially oriented in x-direction which results in a more than 50 % higher thermal conductivity in x-direction than in y-direction. Both effects – influence of the filling factor and of the local orientation of the fibers – are less pronounced in the carbon paper as the filling factor is more homogeneous for the carbon paper (compare Fig. 2) and the local orientation of the fibers is more random.

The length scale of the local variations of thermal conductivity in a carbon cloth based GDL is the length scale of the bundle diameter – typically a few 100 μm – which is comparable to the dimensions of the flow channels in a bipolar plate. Therefore, the orientation and lateral offset of a carbon cloth based GDL relative to the flow field of the bipolar plate might possibly have an influence on fuel cell performance.

The addition of PTFE to the bare carbon paper should lead to an increased thermal conductivity and this is observed. The calculated through plane thermal conductivity of 2.08 W m⁻¹ K⁻¹ of the Toray carbon paper with 30 wt% of PTFE is higher than the values for Toray carbon paper TGP-H-060 without PTFE reported in literature: 1.7 W m⁻¹ K⁻¹ given

by the manufacturer [10], $1.80 \text{ W m}^{-1} \text{ K}^{-1}$ measured experimentally [2] and the range of thermal conductivities of $0.36 - 1.36 \text{ W m}^{-1} \text{ K}^{-1}$ estimated by Ramousse et al. [3] based on experimental data.

Nevertheless it must be considered that in the CT data presented here, the morphology of the contact between the fibers is well defined within the limits set by the resolution of the method. But as PTFE or binder material can not be numerically discriminated from the carbon fibers and the thermal conductivity of all solid material is assumed to correspond to the value for carbon, the thermal conductivity of the junctions between fibers is probably overestimated by the model. It is also not possible to investigate the experimental observation that seems to show that the increased contents of PTFE to a gas diffusion layer leads to a slight reduction in the effective thermal conductivity [2, 3].

The thermal conductivities calculated for different virtual resolutions given in Table 2 show a clear decrease of conductivity with increasing voxel size. This could be expected as for a fiber diameter of approximately $7 \mu\text{m}$, it is reasonable that a voxel size around $1 \mu\text{m}$ is necessary to resolve the fiber structure, while for larger voxel sizes this seems difficult. It seems therefore necessary to use CT with a resolution below $1 \mu\text{m}$ to measure the 3D structure of the fiber network and an even higher resolution will be necessary to correctly determine the structure of the contact regions between fibers. Similarly, it would be desirable to discriminate carbon from PTFE or binder for an even more adequate calculation in particular of the through plane thermal conductivity where thermal conductivity is not dominated by heat transfer along the fibers.

4. Conclusion

Two commercially available gas diffusion layers – the carbon cloth EC-CC1-060T and the carbon paper EC-TP1-060T – were investigated by 3D x-ray computed tomography (CT). Based on the 3D structure reconstructed from tomography data, the macroscopic effective thermal conductivity of the gas diffusion layers was calculated by solving the energy equation considering a pure thermal conduction problem.

The 3D structure of the carbon fibers was clearly resolved by CT, therefore it has been demonstrated that 3D x-ray computed tomography is a suitable technique for the determination of the 3D structure of fiber based gas diffusion layers.

Based on the CT data, the anisotropic thermal conductivity of both samples was calculated. The average in plane thermal conductivity for both samples, the carbon cloth and the carbon paper, is by about a factor 6 to 7 larger than the average through plane thermal conductivity. Furthermore a clear dependence of thermal conductivity on the filling factor and – especially for the carbon cloth – also on the local orientation of the fibers was observed.

Acknowledgment

The authors wish to thank GE Sensing & Inspection Technologies, Wunstorf, Germany for the provision of CT data.

References

- [1] M.F. Mathias, J. Roth, J. Fleming, W. Lehnert, Diffusion Media Materials and Characterisation. Handbook of Fuel Cells – Fundamentals, Technology and Applications, Edited by W. Vielstich, H.A. Gasteiger and A. Lamm, Volume 3 Fuel Cell Technology and Applications, John Wiley, 2003
- [2] M. Khandelwal, M. Mench, Direct Measurement of Trough-Plane Thermal Conductivity and Contact Resistance in Fuel Cell Materials. Journal of Power Sources 161 (2006), 1106-1115
- [3] J. Ramousse, S. Didierjean, O. Lottin, D. Maillet, Estimation of the Effective Thermal Conductivity of Carbon Felts Used as PEMFC Gas Diffusion Layers. International Journal of Thermal Sciences 47 (2008), 1-6
- [4] I. Nitta, O. Himanen, M. Mikkola, Thermal Conductivity and Contact Resistance of Compressed Gas Diffusion Layer of PEM Fuel Cell. Fuel Cells 08 (2008), 111-119
- [5] V.P. Schulz, J. Becker, A. Wiegmann, P.P. Mukherjee, C.Y. Wang, Modeling of Two-Phase Behavior in the Gas Diffusion Medium of PEFCs via Full Morphology Approach. Journal of the Electrochemical Society 154 (2007), B419-B426
- [6] E. Sadeghi, M. Bahrami, N. Djilali, Analytic Determination of the Effective Thermal Conductivity of PEM Fuel Cell Gas Diffusion Layers. Journal of Power Sources 179 (2008), 200-208
- [7] F.H. Garzon, R.L. Borup, J.R. Davey, Micro and Nano X-Ray Tomography of PEM Fuel Cell Membranes after Transient Operation. ECS Transactions 11 (2007), 1139-1149
- [8] P.K. Sinha, P. Halleck, C.Y. Wang, Quantification of Liquid Water Saturation in a PEM Fuel Cell Diffusion Medium Using X-Ray Microtomography. Electrochemical and Solid-State Letters 9 (2006), A344-A348
- [9] P.K. Sinha, P.P. Mukherjee, C.Y. Wang, Impact of GDL Structure and Wettability on Water Management in Polymer Electrolyte Fuel Cells. Journal of Materials Chemistry 17 (2007), 3089-3103
- [10] Toray carbon paper – Manufacturer data sheet. Web: http://www.torayca.com/properties/en/images/report_eng09_2.html
- [11] A. Wiegmann, A. Zemitis, Fraunhofer ITWM Technical report Nr 94 (2006)
- [12] GeoDict, www.geodict.com