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Measurements of GDL Properties for Quality Control in Fuel Cell Mass Production Line

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Summary

National Research Council Canada (NRC) has been reorienting its fuel cell activities towards manufacturing challenges. Some challenges in industrialization of fuel cell manufacturing concern quality control, supplier development and process development. As the gas diffusion layer (GDL) of a proton exchange membrane (PEM) fuel cell plays a critical role in cell performance, NRC has been working closely with Vancouver-based fuel cell companies to develop and validate quality assurance methods to characterize GDL attributes and properties, which strongly correlate with PEM fuel cell performances.

Keywords: PEM fuel cell (proton exchange membrane), electrode, component, commercial, Canada

1 Introduction

Vancouver is a world center in the automotive Fuel Cell industry, which is reflected in the significant advances made in the fuel cell vehicle technology at Automotive Fuel Cell Cooperation (AFCC) and Ballard. Recently Mercedes Benz Canada (MBC) established a mass production line in Vancouver for proton exchange membrane (PEM) fuel cell stack, targeting vehicle applications and signifying the new era of PEM fuel cell technology towards commercialization. As such, NRC has been reorienting its fuel cell activities towards manufacturing challenges. Some challenges in industrialization of manufacturing of fuel cells concern quality control, supplier development and process development.

The GDL of a PEM fuel cell plays a critical role in the cell performance. Desirable functions of an ideal GDL include effectively transporting the reactant gas and removing liquid water, conducting electrons and heat, and having a low contact resistance [1][2]. During manufacturing of a membrane-electrode assembly, MEA, the GDL goes through processing e.g. unwinding, cutting, bending and laminating that will lead to changes in its attributes, which may affect the performance of the resulting MEA in the end. These changes have to be understood in order to properly define specifications to suppliers. To guarantee that the accumulated changes will not exceed the allowable range, quality control and characterization methods will have to be developed. In this context, NRC has been working closely with Vancouver-based fuel cell companies to develop and validate a set of quality assurance methods to characterize GDL attributes and properties, which strongly correlate with PEM fuel cell performances.

Regarding GDL characterization, there have been some reported work in literature; however, not many of them were developed for mass production quality control purposes. Although some of the GDL parameters such as thickness, electrical conductivity, porosity and permeability are measured and provided by the GDL

suppliers, these parameters are only part of the entire spectra of GDL attributes that are critical for ensuring satisfactory performances of the GDLs after the MEA manufacturing processes. Overall, no systematic study of GDL properties for the MEA processing and fuel cell mass production line has been carried out in terms of fundamental understanding of structure and property changes, characterization tools, protocols and standards. As such, this work aims to identify GDL attributes, develop experimental tools for GDL structure and property characterization, validate the design and measurement results, and establish quality assurance protocols and standards based on the knowledge and tools developed.

2 Identification of GDL attributes/properties

2.1 GDL macro component and micro structure [3]

A GDL typically contains the macro-porous gas diffusion backing layer (with a thickness between 170 and 300 μm) and a micro-porous layer (MPL) that has a thickness of between 50 and 100 μm.

The most commonly used materials for gas diffusion backing layer are carbon fibre-based products (the fibres mostly have a diameter around 7 ~ 10 μm), such as carbon cloths made with woven papers, and carbon papers made with non-woven fabrics and carbon papers made with felt/spaghetti fibres, due to their high porosity (≥70%) and good electrical conductivity. The manufacturing of both carbon paper and carbon cloth start with fibre filament formation and stabilization, followed by resin impregnation (PTFE binding for making carbon paper, or weaving for making carbon cloth). Once the carbon paper or cloth is manufactured, bulk treatment of the material with PTFE is needed to increase and stabilize the hydrophobicity. A wide range of PTFE loadings have been used in diffusion media, generally falling between 5 and 30wt% PTFE.

In addition to the bulk hydrophobic treatment with PTFE, the addition of a micro porous layer (MPL) is widely practiced. The MPL consists of carbon or graphite particles mixed with a polymeric binder, usually PTFE. The MPL has a pore size of carbon agglomerates, between 100 and 500 nm, as compared with 10-30 μm pore size for carbon-fibre-paper substrates.

2.2 GDL functions

The primary five key functions of the GDLs are to provide

1) mechanical support for the membrane electrode assembly (MEA)

2) electronic conductivity between the bipolar plates and catalyst layers,

3) heat removal from the MEA towards the coolant channels of the bipolar plates,

4) the mass transfer of reactants (fuel and oxidant)

5) removal of product water [1][2].

There is a design trade-off among these functions. For example, while three of the five key roles of the gas diffusion layer are improved with increased convection, the electrical contact resistivity is apparently low. As for the compressive force, electronic resistivity is substantially reduced by increasing compressive force whereas permeability (and therefore reactant and product mass transport) is reduced as compressive force increases [2].

2.3 GDL attributes and properties [3]

To fulfil the functionalities described above, GDL has to possess several attributes and properties. These properties and attributes can be classified under GDL functionalities. Table 1 lists the identified GDL attributes and properties. The commonly used testing methods for each property and the availability of each testing method are also identified.

Table 1 GDL attributes, properties and experimental measurements [3]

Functionality	**Attribute**	**Property**	**Measurement**		
			Method	**Tool or equipment**	**Destructive or not**
Mechanical	Tensile strength	Tensile strength	Tensile test	Mechanical tester	D
	Bending stiffness	Bending stiffness	Flexural test/3 point bend test	Mechanical tester	D
	Compressibility	Compression	Surface morphology	SEM	
			Pressure vs. thickness	Pneumatic clamps with pressure control	D
Water transport	Diffusivity	Porosity	Mercury porosiometry	Mercury porosiometry	D
		Pore size distribution	Gas adsorption	BET	
			Mercury porosiometry	Mercury porosiometry	D
		Teflon content	Teflon content	TGA	D
		Water vapor diffusivity	Custom	NRC Custom	D
		Contact angle – Internal	Capillary penetration	Fluorescent imaging	
		Contact angle – External	Goniometry	Contact angle analyzer (FTA100)	D
		Fraction of phobic/philic pores	Water/Decane method	Water/Decane method	D
		Thickness	Micrometer	Micrometer	D
	Water permeability	Water permeability	Flow under pressure difference	Specially designed set-up (through plane)	D
Gas transport	Diffusivity	Porosity	Mercury porosiometry	Mercury porosiometry	D
		Pore size distribution	Mercury porosiometry	Mercury porosiometry	D
		O2 diffusivity	Custom	NRC Custom	D
		Thickness	Micrometer	Micrometer	D
	Gas permeability	Gas permeability in the presence of water	Flow under pressure difference	Specially designed set-up (through plane)	D
Electrical conduction	Conductivity (bulk)	Through-plane resistivity	Pneumatic clamp in a specially designed environmental chamber with controlled RH and T and high resolution loadbank		D
		In-plane resistivity	In-house designed cell (under different RH and temperature)		D
	Conductivity (interface)	Contact resistance	samples of varying thicknesses needed	Custom equipment (e.g., SFU)	D
	Surface contact	Surface roughness	Fit	WYKO	D
Heat transport	Heat conductivity	Thermal conductivity	Heat exchange	Variously designed rigs	D

Through the analysis of the processes and based on the experience and knowledge that NRC and MBC teams have, the GDL properties were down-selected (Surface roughness, thickness, and conductivity under compression). It has to be noted that these selected properties are for the first stage of experimental characterization and are chosen as these measurements can be grouped into five categories corresponding to the effects of five processes (unwind, cut, adhesive application, lamination, and sealing) of GDLs. Some of other non-selected properties maybe added later if future tests confirm the necessity.

3 Measurements of selected properties

3.1 Measurements of thickness and conductivity under compression

The GDL thickness and through-plane resistivity/conductivity are measured using a two in one device developed at NRC. The schematic illustration of the two in one device is presented in Figure 1. This device features gold-coated plates, highly sensitive load cell, and automatic control & data acquisition.

For measuring conductivity, the device includes a pair of gold-coated plates that have an effective area of 5 cm^2 to simulate the two electrodes of a PEM fuel cell; an ESPEC SH-241 environmental chamber that can control the temperature between 25 and 90 °C with an accuracy of ±0.3 °C and the RH up to 70% with an accuracy of ±3%; a pneumatic cylinder that can execute a compressing force up to 300 Psi; a DC power supply (6651A – 0~8V DC with up to 50A current) that supplies the current; a Chroma-6300 load bank to control the current (accuracy of ±0.1% for constant current mode -CI); and high accuracy NI-DAQ (resolution<1.5uV) to measure the voltage drop across the GDL. All the current and voltage data are logged into a computer via NI DAQ using Labview software.

For measuring GDL thickness, a high resolution micro-meter (1 μm) is mounted to the lower part of the gold plated plate so that the distance between the two gold-coated plates, which is the thickness of the GDL held in between the two plates, can be measured.

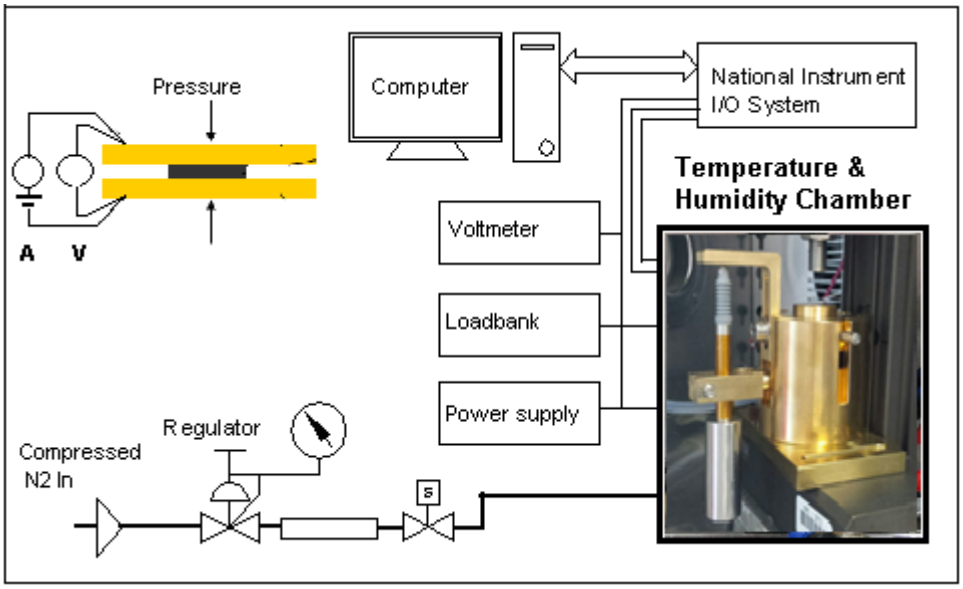
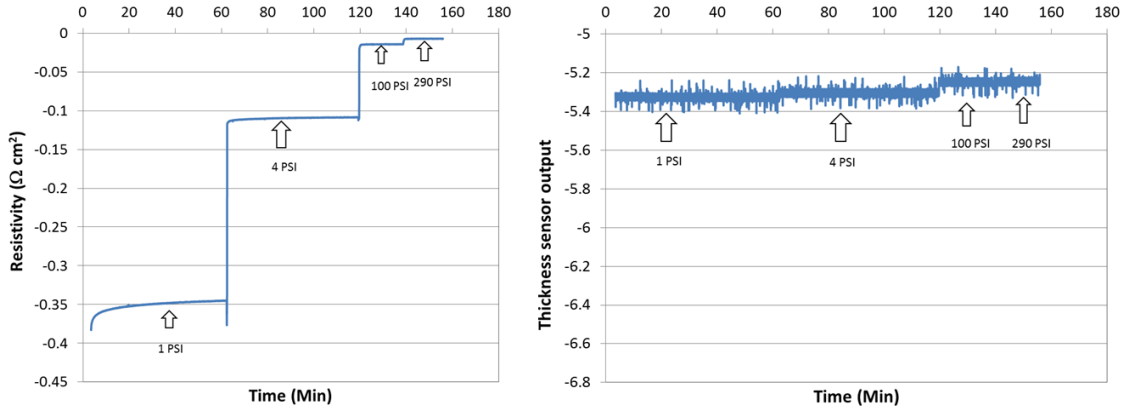


Figure 1 Schematic of the thickness/resistivity measurement and data acquisition system using the two in one device

Sample resistivity output and thickness sensor output curves vs. pressure are given in Figure 2. From these curves, thickness and conductivity under different compressions can then be derived.



(a) (b)

Figure 2 Sample resistivity output and thickness sensor output for (a) Resistivity vs. pressure and (b) Thickness sensor output vs. pressure

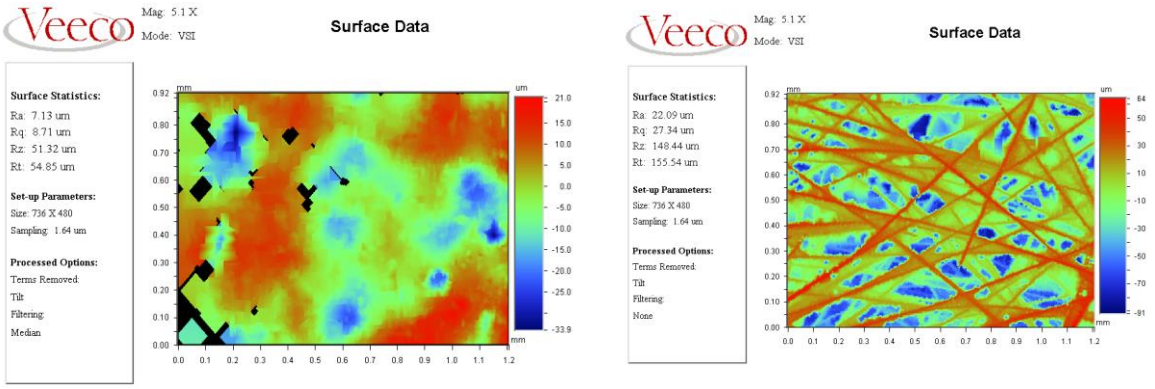
A testing protocol for measuring both GDL thickness and resistivity/conductivity has been established, including sample cutting & placing, test running, compressive force applying, data acquisition, pressure releasing, and data processing.

3.2 Measurements of surface roughness

The WYKO NT 2000 profiler uses non-contacting vertical scanning interferometry technique to process fringe modulation data from the intensity signal for surface height calculations as it vertically scans through a specified distance. Figure 3 shows the picture of the WYKO system, which has a vertical resolution of 3 nm and a spatial resolution of 1.64 μm at a 5x optical magnification.

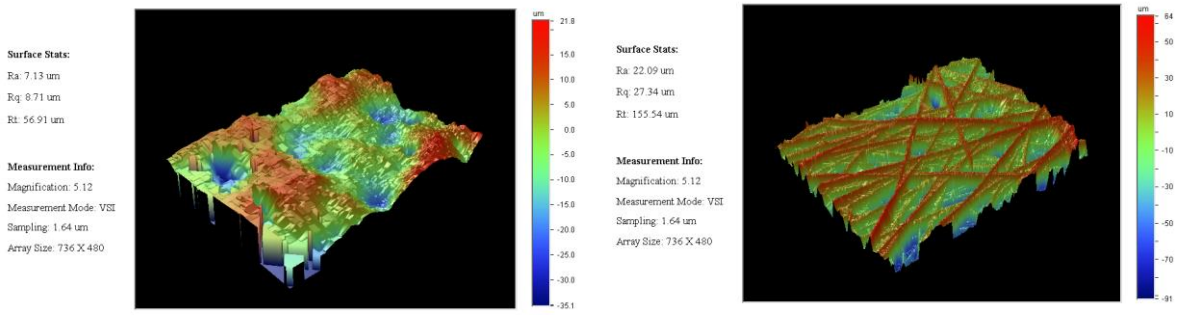
Figure 3 Picture of the optical surface profiler (WYKO) system

For each GDL sample, three areas/spots are measured for surface roughness. In this work, direct measurement rather than imprint is used, therefore, each measuring spot has a dimension of 0.9 x 1.2 mm^2 rather than 4 x 5 mm^2 for imprint measurement. Figure 4 shows examples of typical surface data and Figure 5 shows typical 3D images of measured spots that have an array of 736 x 480. Based on the WYKO optical surface profiler, a testing protocol for measuring GDL surface roughness has been established, including sample cutting, sample placing, focus adjusting, and data acquisition & analysis.



(a) (b)

Figure 4 Typical 2D surface data of a sample GDL on (a) the MPL side and (b) the substrate side of 25BC



(a) (b)

Figure 5 Typical 3D interactive displays of one measured spot that has an array of 736 x 480 on (a) the MPL side and (b) the substrate side of 25BC

4 Results

To benchmark the equipment and its setups, surface roughness and thickness/conductivity under compression are measured using commercial Sigracet® 25BC GDLs.

4.1 Thickness/conductivity under compression

Figure 6 depicts the results of 25BC for both thickness and conductivity measurements with error bars presented. The curves show that conductivity increases with compression while thickness decreases with compression. After a compression pressure of 100 psi, the sample thickness tends to remain relatively stable whereas the sample conductivity keeps increasing.

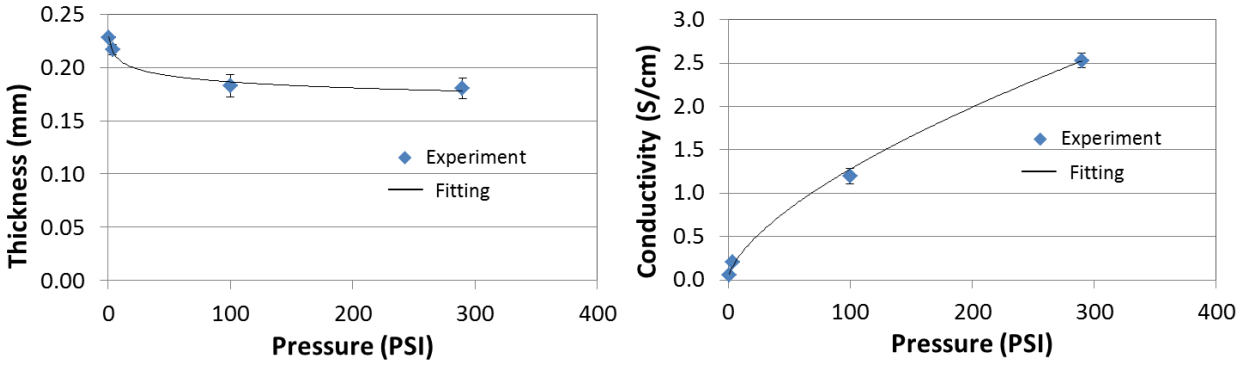


Figure 6 Thickness and conductivity curves vs. compression for 25BC

4.2 Surface roughness

Tables 2 and 3 list the measurement results of surface roughness for pristine and compressed samples on both sides of 25BC. The compressed samples are obtained after thickness/conductivity measurements with a final compression pressure of 290 psi. Note that three samples are measured and each sample is measured with three scans at three different areas/spots.

Table 2 Measurement results of surface roughness for pristine and compressed samples of the MPL side of 25BC (μm)

Sample ID	Ra*	Ra Ave*.	STD*	Rz*	Rz Ave.	STD	Rq*	Rq Ave.	STD
Pristine	5.41	6.11	1.05	10.92	12.63	2.21	6.55	7.54	1.32
	6.60			13.64			8.36		
	6.32			13.35			7.72		
Compressed	5.51	5.32	0.67	10.73	12.30	1.69	6.76	6.70	0.84
	5.68			12.65			7.12		
	4.77			13.51			6.21		

* Ave. = Average; STD = Standard Deviation; Ra, roughness average, is the mean height as calculated over the entire measured array; Rq, root mean square roughness, is the root mean square average of the measured height deviations taken within the evaluation length or area and measured from the mean linear surface. Rz, average maximum height of the profile, is the average of the successive values of Rti calculated over the evaluation length. Rti is the vertical distance between the highest and lowest points of the profile within a sampling length. It is the average of the greatest peak-to-valleys separations.

Table 3 Measurement results of surface roughness for pristine and compressed samples of the substrate side of 25BC (μm)

Sample ID	Ra	Ra Ave.	STD	Rz	Rz Ave.	STD	Rq	Rq Ave.	STD
Pristine	20.15	20.32	2.16	72.24	76.51	9.35	24.59	25.07	2.63
	20.71			82.69			25.43		
	20.11			74.60			25.18		
Compressed	18.46	18.57	2.38	74.28	71.77	9.01	23.52	23.57	2.79
	18.02			66.64			22.76		
	19.24			74.40			24.43		

The parameters from Tables 2 and 3 are also plotted in Figures 7 and 8 with error bars presented. The results suggest that the substrate side is much rougher than the MPL side of the 25BC GDL for both pristine and compressed samples and for all the selected parameters of roughness.

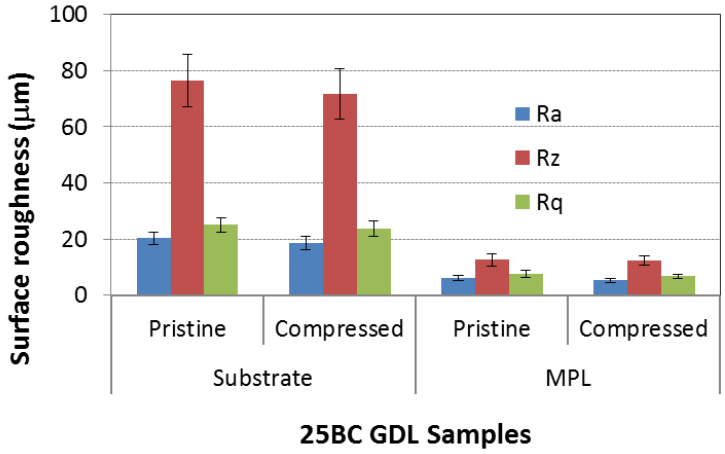


Figure 7 Comparison of surface roughness for pristine and compressed samples of 25BC (Ra vs. Rz & Rq)

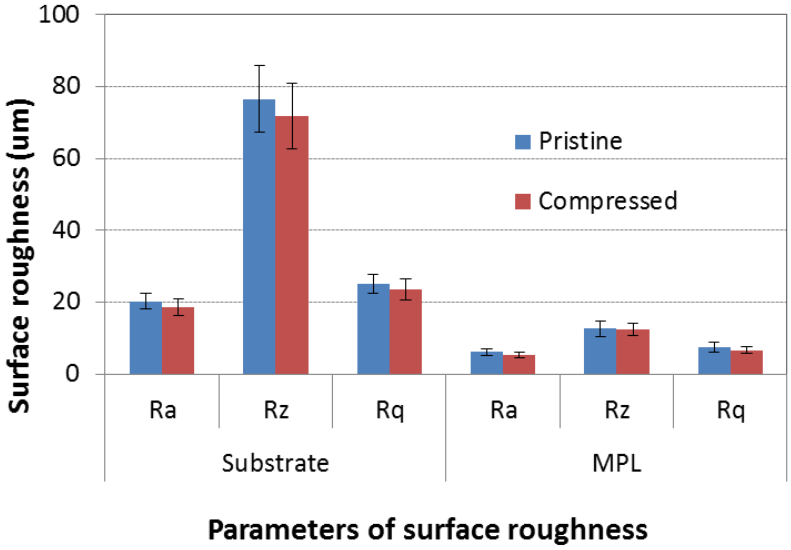


Figure 8 Comparison of surface roughness for pristine and compressed samples of 25BC (pristine vs. compressed)

Using Ra as an example, the MPL side of 25BC has a standard deviation of 1.05 for the pristine samples while the compressed sample has a standard deviation of 0.67. For the substrate side of 25BC, a standard deviation of 2.16 is obtained for the pristine samples while a standard deviation of 2.38 is observed for the compressed samples. A standard F-Test suggests that the standard deviations are not significantly different for pristine vs compressed or substrate vs MPL.

The Ra, Rz and Rq of the compressed samples are less than those of the pristine samples for MPL suggesting a possible reduction in surface roughness of MPL from compression, but a T-Test suggests that there is no statistical significant difference based on a 95% confidence. Similarly, the Ra, Rz and Rq of the compressed samples are also less than those of the pristine samples for substrate suggesting a possible reduction in surface roughness of substrate from compression. However, a T-Test suggests that there is no statistical significant difference based on a 95% confidence for Rz and Rq, while it does suggest that there is a decrease in surface roughness of Ra after compression on the substrate side.

Overall, there is significant decrease in surface roughness of the MPL vs substrate but there is no difference in the standard deviations of MPL surface roughness vs substrate surface roughness. Compression appears to decrease Ra on substrate but has no significant effect on the MPL.

5 Summary

Surface roughness, thickness and conductivity under compression are down selected as the three properties are important to GDL design and manufacturing.

The WYKO surface profiler has been identified and its testing protocols are established for GDL surface roughness measurements. Using WYKO surface roughness is measured using a commercial 25BC GDL. The results show that the substrate is rougher than the MPL side of the 25BC GDL for both pristine and compressed samples. For both substrate and MPL sides of the sample, compressed samples appear smoother than the as unprocessed ones.

An in-house two in one device was designed for thickness/conductivity measurements under compression with testing protocols established. The thickness and conductivity are also measured using a commercial 25BC GDL. In general, conductivity increases with compression while thickness decreases with compression.

In the process of identifying GDL attributes, characterizing GDL properties, and establishing quality assurance protocols, the knowledge and tools are developed, contributing to the specifications definition and quality control of fuel cell components for mass production.

Acknowledgments

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