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Development of Standard Methods and Devices for Measuring GDL Properties

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Summary

Fuel cell technology industrialization, to be successful, will need a strong and well-developed supply chain. Standardization of components specifications measurements is an essential part to develop a supplier base, maximizing quality control to reduce cost. Among the various components, the gas diffusion layer (GDL) is a complex material fulfilling several functions to ensure proper performance of a fuel cell. This work aims to develop a three-in-one device to characterize GDL attributes and properties, including thickness under compression (TUC), resistivity under compression (RUC), and in-plane permeability (IPP) under compression. The idea of a TUC/RUC device is not new with commercially available product. Adding IPP to the TUC/RUC device is quite challenging. This can be realized by measuring the flow rate–pressure relationship in a radial-flow apparatus through an annulus of GDL material. As such, a TUC/RUC/IPP tester is developed, featuring simultaneous measurements of TUC, RUC and IPP under compression in one device with automated data acquisition.

Keywords: fuel cells; supply chain

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1 Introduction

As industry is commercializing fuel cell technologies, especially in transportation applications, there are needs and challenges in the industrialization of manufacturing that includes quality control, supplier and process development for several components including the GDL.

1.1 GDL

GDL is a crucial component in the Proton exchange membrane fuel cell (PEMFC) stack. 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].

The GDL typically consists of a macroporous carbon cloth or carbon paper substrate that is optionally coated with a thin microporous layer (MPL) made of carbon black. Carbon fibres of the substrate are graphitized at high temperature (>2000 °C) to enhance electronic conductivity, corrosion resistance and mechanical strength, and impregnated with thermoset resin to manufacture carbon papers. The GDL is typically wet-proofed with polytetrafluoroethylene (PTFE) to prevent liquid water from clogging into the pores, which could impede gas transport within the GDL [1].

Major GDL properties include mechanical properties, (e.g. compressibility, tensile properties, and bending), porosity, permeability (including air permeability and water vapour permeability), polytetrafluoroethylene (PTFE) content, electric conductivity (including in-plane and through plane conductivity), thermal conductivity [1], and surface roughness. An ideal GDL should offer properties such as superior gas diffusion with optimum bending stiffness, porosity, surface contact angle, air permeability, water vapour diffusion, hydrophobicity, hydrophilicity, corrosion resistance, crack-free surface morphology, high mechanical integrity and enhanced oxidative stability along with durability at various operating conditions including freezing. Understanding the design and functional characteristics of GDL may provide a significant contribution in the gas diffusion process and components to optimize the quality of MEAs [3].

In support of commercializing the fuel cell technology, National Research Council Canada (NRC) has established an open group/consortium including fuel cell manufacturers, test equipment suppliers and GDL suppliers to tackle specific challenges linked to the GDL supply chain, specifically on the standardization of specifications measurements and quality control.

1.2 TUC, RUC, and IPP

The accurate measurement of thickness is crucial for calculating the bulk volume of thin porous materials such as GDLs [4], and accurately gauging sample thickness is not trivial. The difficulties of measuring GDL thickness arises from GDLs' small and non-uniform thickness, especially under compression. Because the material thickness can depend heavily on the compressive force, the pressure used should be reported when reporting the thickness. The size of the gauge foot and the length of time under compression before measurement is taken should also be controlled for consistent results [5].

The resistance is normally a through-plane sheet resistance. This property is typically measured by putting a sheet of diffusion media between two flat plates, applying a defined compression, applying a d.c. current through the material, and measuring the plate-to-plate voltage drop. In general, the measurement techniques discussed in literature can be categorized into two groups: 2-point probe method and 4-point probe method. This measured resistance includes contributions from the bulk material and the two contact resistances between diffusion media and plates. In attempts to isolate the bulk resistance, gold plates or even mercury contacts can be used to minimize the contact resistance [5].

One of the main parameters that would improve the overall performance of a fuel cell is better mass transport of reactants through the diffusion layer (DL) toward the active catalyst zones. To quantify and characterize how well the gas mass transport is in a specific DL material and design, it is important to measure the in-plane and through-plane permeability [6]. The need for reliable methods to measure gas diffusion layer permeability at various compressive loadings has been identified by the fuel cell community as critical to develop. In a recent review on gas diffusion layer characterization, Mathias et al. [5] points to in-plane permeability as the relevant parameter in fuel cell performance, citing diffusion as the dominant mechanism for through-plane transport. The view that in-plane permeability should be more relevant has been reinforced analytically by Feser et al. [7] as well as numerically by Pharaoh [8], particularly with respect to PEM fuel cells that employ serpentine flow fields. Further, transport of reactants in the through plane direction is a diffusive mechanism rather than a convective mechanism; thus, the only pressure driven mass transfer occurs in the in-plane direction. Several theoretical and experimental studies also have shown that the in-plane permeability of GDLs is a key parameter in optimization of PEMFCs' performance [9]. However, transverse or through the thickness (through-plane) permeability is most commonly measured, whereas fewer techniques have been used to characterize the in-plane permeability of the gas diffusion layer [2]. So far, no standard, generally accepted permeability test has been established.

Through discussions and ranking within the project team, three properties: (1) In-plane permeability (IPP), (2) Thickness under compression (TUC), and (3) Resistivity under compression (RUC) (through plane), have been selected with the goal of identifying the key properties for developing standard measuring methods and devices. It is believed that TUC, RUC, and IPP are the most suitable and useful properties for developing standardized devices and testing protocols for the GDL supply chain.

Thermal conductivity, due to the foreseeable lengthy time needed for reaching steady-state during the measurement, might not be a practical property for quality control. Surface roughness, due to the complexity and high accuracy requirements associated with the nano-scale measurement, needs more debate on whether it would be a good quality control tool. As a result, the scope of this work does not include the tool development for thermal conductivity and surface roughness.

This work aims to develop a three-in-one device, which is able to measure IPP, TUC, and RUC using the same device. Instead of individual device to measure each property, this device measures multiple properties and carries out property characterizations in a timely and cost-effective manner. In particular, this experimental tool is beneficial for the quality control of mass production. Further targets are to establish quality assurance protocols and standards.

2 Design requirements

Measurements of TUC and RUC are basically straightforward, and TUC/RUC testers are commercially available. One of the challenges for developing a TUC/RUC/IPP tester is to add IPP to the TUC/RUC device. This can be realized by measuring the flow rate–pressure relationship in a radial-flow apparatus through an annulus of GDL material. Another challenge could be achieving the necessary sealing, which can be alleviated by constraining the upper and lower surfaces of the annulus between two sufficiently flat and smooth plates. The main critical performance requirements for the device are summarized in Table 1.

Table 1: Critical Design Requirements

Characteristics		Range	Accuracy	Resolution
TUC		0-1000 μm	± 1 μm	< 1μm
RUC		10 μΩ-100 Ω	± 0.5%	< 2 μΩ
IPP	Flow rate	1-1000 sccm	± 2%	< 2 sccm
	Pressure	1-100 kPa	± 1%	< 50 Pa
Compression		0-5/10 MPa	± 1%	< 20 kPa
Plate flatness		Maximum unevenness of 1 μm		

To meet the performance requirements of the TUC/RUC/IPP Tester, great efforts have been made to develop the three-in-one device. The design of the device is based on the previous Greenlight TUC/RUC tester and Automotive Fuel Cell Cooperation (AFCC)'s TUC/RUC/IPP tester (Daimler Fuel Cell Research design) along with the experience from the consortium members. For the ease of use, manufacturability, equipment durability, cost, and setup/maintenance time, the design is carefully optimized. The schematic drawings of the process from a TUC/RUC tester to a three-in-one device for TUC/RUC/IPP measurements are shown in Figure 1.

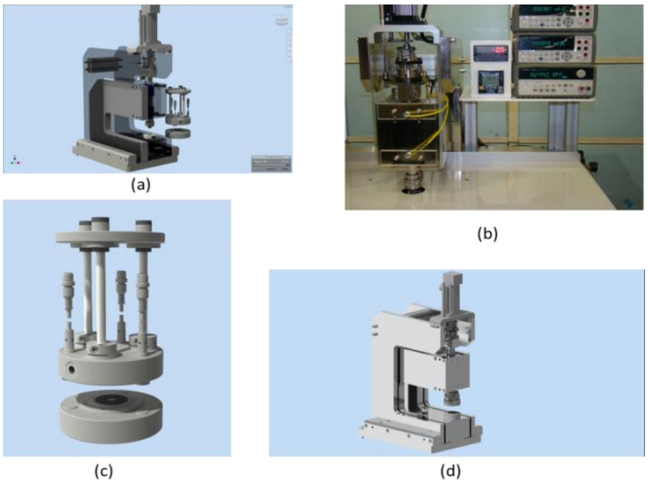

Figure 1: The schematic drawing and images of the three-in-one device for TUC/RUC/IPP measurements

(a) TUC/RUC head (b) Image of the TUC/RUC device (c) Standalone IPP head (d) TUC/RUC/IPP measuring head

3 Results and discussion

3.1 Features of the device

Figure 2 shows the image of the finished TUC/RUC/IPP device. The method of compression uses an electro-pneumatic regulator controlled pneumatic cylinder, with a load cell in series, feeding back the force to the electro-pneumatic regulator. The specifications on the TUC/RUC/IPP tester include repeatable thickness measurement of 0-1mm with a repeatability of ±1 microns under a varying load up to 5/10 MPa. Resistivity measurements are possible from micro-ohms to mega-ohms, with an accuracy within ±0.5%. The integrated IPP functionality measures in-plane permeability with an accuracy of ± 2%. With the help of MATLAB, a LabVIEW based interface was designed and developed for control of the TUC/RUC/IPP tester. What makes this design suitable for a production tester is its automated data collection ability, 2 handed operation for safety, pneumatic controlled and calibrated sample force, and the ability to do pre-programmed routines (varying pressure, current, force and airflow).

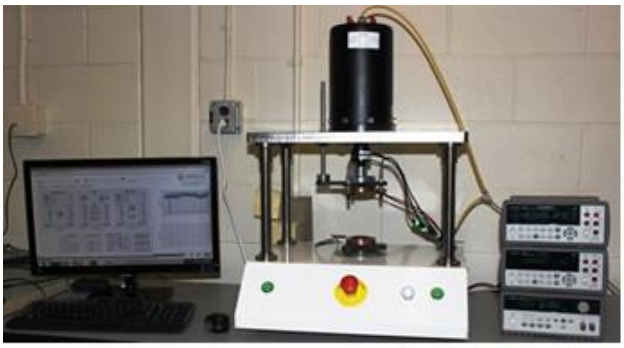

Figure2: Image of the finished TUC/RUC/IPP device

3.2 Calculations from measurements

The key of this device design is to use an annular GDL specimen. Using the variables presented in Figure 3, resistivity, conductivity and permeability can then be calculated.

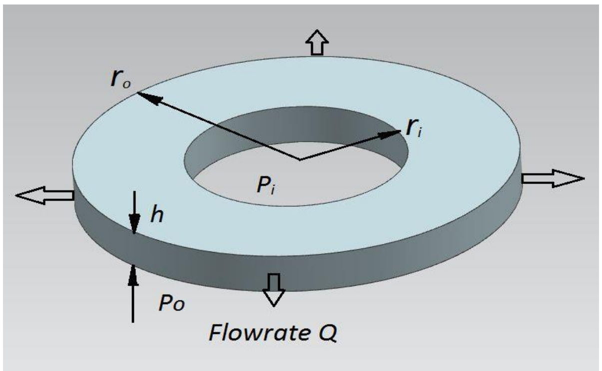

Figure3: Variables used for the calculation

Calculation of resistivity (ρ) and conductivity (σ):
The Current and Voltage applied across the annular sample are measured in addition to the thickness. Combined with a known cross sectional area, resistivity (unit: *Ω·m*) is measured using the following equation:

$$\rho = \frac{V}{I}\frac{A}{h} \tag{1}$$

where

V is the voltage applied across the donut-shaped GDL sample (*V*)
I is the current applied across the donut-shaped GDL sample (*A*)
h is the thickness of the GDL sample (*m*)
A is the cross-sectional area of the GDL sample (for a donut-shaped sample, $A = \pi(r_0^2 - r_i^2)$) (m^2)

Conductivity, σ (*S/m*), is defined as the inverse of resistivity:

$$\sigma = \frac{1}{\rho} \tag{2}$$

Based on Darcy's Law under the assumption that in-plane permeability is homogenous and transversely isotropic, in-plane permeability (k_i, m^2) can be calculated as follows. Note that an often used permeability unit is Darcy. One Darcy is equivalent to 9.87×10^{-13} m² or 0.99 (μm)².

$$k_i = \frac{\mu Q}{2\pi h \Delta P} ln \frac{r_0}{r_i} \tag{3}$$

where

μ is the fluid viscosity ($Pa{\cdot}s$)
Q is the measured flow rate (m^3/s)
h is the thickness of the GDL sample (*m*)
ΔP is the difference of the inner pressure and the outer pressure ($\Delta P = P_i - P_o$, Pa)
r_o and r_i are, respectively, the outer and inner radius of the annular GDL sample (*m*)

3.3 Equipment Functions

The functions of the device include:

1. TUC & Compression Measurement
    - Triangulated contact probes to measure thickness and check platen tilt (parallelism)
    - Screen display of set points and actual thickness and pressure values
    - List of pre-set set points to improve data acquisition time.
2. RUC Measurement & Control
    - Settable to specified current & voltage across sample
    - Settable direction of current flow (Top to bottom or vice versa)
    - Screen display of set points and measured values
3. IPP Measurement & Control
    - Pressure set and mass flow measurement through sample
    - Mass flow rate set and pressure measurement across sample
    - Screen display of set points and actual pressure and mass flow data
    - List of pre-set set points for pressure and mass flow to speed data acquisition time.
4. Automated Data Collection
    - Display of the current sample reading on screen
    - Automatic data logging to a test results file for further analysis
    - Control to modify author, data descriptions, titles and identifiers during test

4 Summary

NRC has reoriented its activities over the last 5 years from new materials development towards fuel cell manufacturing challenges in support of the industry commercialization efforts of fuel cell technology, especially in transportation applications. Some challenges in industrialization of fuel cell manufacturing concern quality control, suppliers development and process development. NRC has been working closely with fuel cell manufacturers, fuel cell testing equipment manufacturers, and fuel cell component suppliers to tackle some of these challenges.

GDL in a PEM fuel cell plays a critical role in cell performance. To characterize GDL properties, TUC, RUC and IPP under compression have been selected for developing standard measuring methods and devices. The designed TUC/RUC/IPP testing station is a bench-top precision instrument capable of providing accurate and precise control, automated testing protocols, loading customized testing scripts, exporting or displaying the measurement data, and simultaneously conducting TUC, RUC and IPP measurements of a gas diffusion layer in a consolidated platform. Unfortunately, the manufacturing of the device has just been completed. The validation of this three-in-one device is still in progress. The next step will be to conduct TUC/RUC/IPP measurements using commercial GDL samples with a variety of thicknesses and properties. Standard procedures and protocols will also be developed.

This work is accomplished through close collaborations between NRC laboratories, and members of the fuel cell industrial sector in Canada and internationally. NRC has brought fuel cell producers, equipment manufacturers and component suppliers together to tackle challenges towards industrialization of fuel cell manufacturing, specifically, to develop standardized methods, protocols and hardware for the measurement of GDL specifications. This project contributes to the specifications definition and quality control of GDL for fuel cell mass production.

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