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Applying infrared thermography as a quality-control tool for the rapid detection of polymer-electrolyte-membrane-fuel-cell catalyst-layer-thickness variations

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ABSTRACT

As fuel cells become more prominent, new manufacturing and production methods are needed to enable increased volumes with high quality. One necessary component of this industrial growth will be the accurate measurement of the variability of a wide range of material properties during the manufacturing process. In this study, a method to detect defects in fuel cell catalyst layers is investigated through experiment and mathematical simulation. The method uses infrared thermography and direct-current electronic-excitation methods to detect variations in platinum-containing catalyst-layer thickness with high spatial and temporal resolution. Data analysis, operating-condition impacts, and detection limits are explored, showing the measurement of defects on the millimeter length scale. Overall, the experimental and modeling results demonstrate great potential of this technique as a nondestructive method to measure defects that is amenable to use on roll-to-roll manufacturing lines.

1. Introduction

Materials and systems advances continue to improve the performance and durability of polymer electrolyte membrane fuel cell (PEMFC) systems, but cost continues to be a barrier to wider adoption [1]. However, PEMFC systems are currently being deployed in some early markets, and as this commercialization expands, there must be a transition from low-volume, largely manual manufacturing methods, to high-volume, continuous, and automated processes. In parallel, quality-control techniques must be developed that are sufficiently sensitive, rapid, noncontact, and nondestructive, and that can be implemented in continuous or automated production lines to inspect the entire material. Scalable quality-control methods are especially needed for membrane electrode assembly (MEA) components, including membranes, electrodes, and gas diffusion media (GDM), which are often inspected visually before use in a stack [2]. Of particular importance, given the high cost of Pt and other noble metal constituents, is the detection of variations in catalyst layers. Catalyst layer thickness variations, such as a spot with excessive or limited loading, can yield performance losses beyond those expected from the Pt-catalyst variation alone, especially at the local area near the defect [3–5]. This is most likely due to uneven compression of the GDM at the location of the defect, which increases resistance and leads to water management and other issues [5].

Today many MEA components are fabricated using roll-to-roll processes [2], wherein a sheet of material that is continuously formed by coating, casting, or other methods is conveyed through required process steps by a roller system, and wherein the final product is wound into a continuous roll (see [6,7] for details on general roll-to-roll processing). A large existing industry provides in-line inspection devices for typical roll-to-roll products such as adhesive tape and paper towels. These devices typically concentrate on optical inspection of visible defects or point measurements of coating thickness. Although these measurements can be extremely useful, they cannot necessarily be applied to PEMFC MEA components, because (1) not all defects are visible, particularly relative to the typically black, nonreflecting electrode coatings and GDMs; and (2) point measurements provide only statistical data – they generally cannot locate discrete defects, even if scanned across the sheet of material being fabricated, as is often done in the industry. Given

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that discrete defects can act as failure-initiation points for MEAs in operation [8], an inspection technique must be developed that measures every location on the sheet material such that defects in MEA components can be removed prior to assembly into complete cells. This 100% inspection requirement is necessary to avoid premature cell failures, which often can cascade and cause complete stack failure and necessitate expensive stack replacement.

For catalyst layer defects, X-ray fluorescence (XRF) [9,10] has recently been applied to the in-line measurement of catalyst loading [11]. Unfortunately, this technique cannot at this time provide 100% inspection for catalyst layer defects. Acquisition times provided by commercially available systems to provide for quantitative analysis of Pt loading are on the order of 10 ms [12]. At these acquisition times, assuming a roll width of 18 in. and a web speed of 30 ft per minute, data are gathered only every 1.42 ft in the direction of substrate motion. Therefore, this technique can provide only a statistical metric of Pt.

Infrared (IR) thermography may enable complete, rapid (with response times on the order of 1 s for large areas), noncontact, and nondestructive detection of defects. In fact, several patents mention using IR thermography to detect various PEMFC parameters, including MEA and membrane defects [13–17]. However, little public information is available about the ability of specific excitation techniques to enable the detection of specific (size and type) defects in PEMFC materials. Although IR thermography techniques may not be element specific (as is XRF), this work shows that IR techniques can be used to identify catalyst layer thickness variations rapidly, at a relatively low cost, and with 100% inspection.

In this paper, a DC-based excitation technique is examined experimentally and theoretically for detecting variations in catalyst layer thickness via IR thermography (henceforth, IR/DC). As an initial study in the use of IR/DC detection for catalyst layer defects, this work focuses on detecting square spots of different thicknesses in a surrounding layer of nominal thickness. Assuming that Pt is uniformly distributed on the carbon support in the catalyst layer, this method allows for the quantification of the areal Pt loading using the known Pt/C ratio. Section 2 describes the experiment and theory for the technique. Section 3 presents the results of IR measurements with defined defects that are used for model validation. Section 4 uses experiments and modeling to examine the impact of different variables of the technique, including excitation time and voltage, and defect size and aspect ratio, as well as fuel cell testing data to verify that the technique does not damage the electrode material.

2. Experimental

Catalyst layer samples were prepared from a catalyst ink containing Ion Power Inc. DE2020 solution (20 wt% 1100 EW Nafion), n-propanol, water, and TKK Inc. TEC10E50E 46% Pt/Ketjen carbon catalyst. The ionomer to carbon mass ratio was 8:10. The total solids content in the ink was 6.8%. All components were hand mixed with a glass stir rod in a glass vial, sonicated for 4 h (in an ice bath), hand mixed, transferred to a 20-mL syringe, and horn sonicated for 30 s. The catalyst ink was then sprayed onto a 150-μm thick, 5-cm × 5-cm ethylene tetrafluoroethylene (ETFE) decal using a home-built ultrasonic spray system utilizing Sono-Tek's impact line of spray heads [18]. The spray system includes a syringe pump, a 120-kHz micro-bore spray head and power system, and a programmable x–y stage. Details of optimal conditions based on spray material can be found in [18]. The flow rates of the catalyst ink during spraying and the spray head conditions were optimized for the system to achieve a Pt loading of about 0.04 mg cm^{-2} in a single coat. The spray process was performed on a heated stage at 80 °C. No additional time for drying was needed between coats. Defects were made by applying a removable polymeric mask over a square region (typically 1 cm × 1 cm) in the center of the 25-cm^2 layer. The mask was removed during the deposition process to achieve the desired (lower) defect area thickness (loading). No residual adhesive remained after the mask was removed.

Fig. 1 shows schematics of the experimental setup and the experiment. Unless stated otherwise, a 21-V DC bias was applied to the setup using a Keithly 2400 source. For alternative voltages a BK Precision XLN 10014 Power Supply was used. Catalyst layer samples on the ETFE decals were fit between two brass clamps, which were clamped with two bolts torqued to 10 cm-kg$_f$. To detect the temperature variation, an IR camera (Jenoptik Vario-Cam HiRes) was used. The camera has a 640 × 480 pixels detector with accuracy under ±1.5 K. A uniform emissivity of 0.95 was used and was determined empirically by comparing a piece of 3 M brand masking tape, which has a known emissivity of 0.95 [19], at the same ambient conditions as the catalyst layer.

For cell testing, the defect-containing decals were hot-pressed onto Nafion 212 membranes at 130 °C under 120 lb$_f$-cm$^2_{\text{active area}}$ pressure for 5 min and assembled into cells using SGL25BC GDLs. Performance curves conducted after break-in were run with hydrogen and oxygen utilizations of 83% and 50% (1.2 and 2 stoich), respectively. Forward and then backward scans of the current from 0 to 996 mA cm^{-2} were executed in 6 increments with 15-min current holds at each point along the curve. The last 5 min of each data point were averaged for reporting.

3. Modeling

The development and analysis of the diagnostic technique was aided by mathematical modeling. For the computational models, the governing heat conduction and current conservation equations are:

$$\rho_i c_{p,i}\left(\frac{\partial T}{\partial t}\right) - \nabla\cdot(k_i \nabla T) = Q \tag{1}$$

and

$$-\nabla\cdot(\sigma_i \nabla \Phi) = 0 \tag{2}$$

respectively, where ρ_i, $c_{\mathrm{p},i}$, σ_i, and k_i are the density, specific heat capacity, electronic conductivity, and thermal conductivity of species i, respectively, T is the absolute temperature, t is time, and Φ is the electronic potential. The Joule heating rate per unit volume is computed from the dot product of the electric field and current density, $\boldsymbol{i}$,

$$Q_i = -\boldsymbol{i}\cdot\nabla\Phi \tag{3}$$

where the subscripts i in Eqs. (1)–(3) can be catalyst layer, ETFE decal, or catalyst layer defect.

The boundary conditions for the model are:

$$T = T_{\text{room}} \text{ and } V = V_{\text{applied}} \quad \text{for } \partial\Omega \in x = 0 \tag{4}$$

$$T = T_{\text{room}} \text{ and } V = 0 \quad \text{for } \partial\Omega \in x = w \tag{5}$$

$$n\cdot(-k_i \nabla T) = h_{\text{side}}(T - T_{\text{room}}) \quad \text{for } \partial\Omega \in y = 0 \text{ and } d \tag{6}$$

$$n\cdot \mathrm{J} = 0 \quad \text{for } \partial\Omega \in y = 0 \text{ and } d \tag{7}$$

$$n\cdot(-k_{\text{ETFE}} \nabla T) = h_{\text{bottom}}(T - T_{\text{room}}) \quad \text{for } \partial\Omega \in z = 0 \tag{8}$$

$$n\cdot(-k_{\text{CL}} \nabla T) = h_{\text{top}}(T - T_{\text{room}}) \quad \text{for } \partial\Omega \in z = h \tag{9}$$

where w, d, h are the width, depth, and height of the computational domain, respectively, and h_{side}, h_{top}, h_{bottom} are the convective heat transfer coefficients for the side, top, and bottom walls, respectively. The parameter values used in the numerical simulations are listed in Table 1, where the convection heat transfer coefficients are