INFRARED THERMOGRAPHIC DEFECT DETECTION IN FUEL CELL GAS DIFFUSION ELECTRODES

By

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ABSTRACT

Polymer Electrolyte Membrane fuel cells (PEMFCs) are energy conversion devices that offer high power densities and high efficiencies for mobile and other applications. Successful introduction into the marketplace requires addressing cost barriers such as production volumes and platinum content. The platinum-catalyst employed in PEMFC electrodes is a primary cost driver for manufacturing. For cost reduction, it is vital to minimize waste during large-scale production of electrodes, including gas diffusion electrodes (GDEs), by developing quality control (QC) diagnostics suitable for a continuous manufacturing environment. In this work, the development of an infrared thermography QC diagnostic for a GDE manufacturing web-line is conducted. A non-flammable H₂/O₂ gas mixture in N₂ was passed through the GDE, reacting exothermically with the platinum catalyst, causing the GDE temperature to rise. Infrared imaging of the variations in the GDE’s thermal profile revealed manufacturing defects and non-uniformities in the catalyst loading. Experiments with a moving substrate were conducted to demonstrate the applicability of the diagnostic for real-time web-line inspection.

Initial experiments used a stationary enclosed testing manifold made of brass to quantify the thermal response caused by the gas-catalyst reaction. This manifold allowed for a controlled environment and to force all gas through the GDE. Experiments with the stationary manifold demonstrated successful detection of GDE defects. The GDE was then suspended above the manifold to simulate open-air testing. Although the thermal response decreased with increased offset distance, successful defect detection was demonstrated. Next, a perforated-tube gas knife was fabricated to deliver a uniform gas flow across the GDE surface in a line pattern. The gas knife uniformity was tested by microscope inspection of the gas knife holes and a hot plate cooling experiment. Finally, the diagnostic was demonstrated on a moving GDE by constructing a bench top roller system with a height and angle-adjustable gas knife holder. The GDE thermal response was tested under varying gas knife height, angle, flow-rate, and hydrogen concentrations. The optimized QC diagnostic was then demonstrated under conditions typical of a manufacturing environment, resulting in the successful detection of a 2 mm square defect on a GDE moving at 30 feet per minute. These results indicate that this QC diagnostic for detecting defects on GDEs is effective.
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CHAPTER 1 INTRODUCTION

There is a growing need to use non-petroleum based energy sources in order to protect the environment and increase energy security [32]. Fuel cells are promising devices that efficiently convert chemical energy (fuel and oxidant) to electric potential energy. Polymer electrolyte membrane fuel cells (PEMFCs) convert hydrogen and oxygen via catalytic reactions at an anode and cathode respectively into electric energy. Research shows that while super-capacitors, batteries, and flywheels are effective energy storage devices, fuel cells are a prime candidate for energy conversion because of relatively high efficiencies (estimated around 50%) in addition to the ability to be implemented along side methane or methanol production [32]. Further, when operated on hydrogen, fuel cells can operate with water as the only exhaust byproduct.

The broad adoption of fuel cells is largely dependent on cost reduction of materials and mass production of materials. Electrodes, which contain platinum (Pt) to catalyze the reaction, are a key component of PEMFCs. Their functionality is vital for reliable operation of the fuel cell system. Quality control of material functionality and homogeneity during the production process is a means to reduce cost and to ensure high reliability prior to stack assembly. Such diagnostics need to be rapid, non-destructive, relatively inexpensive, and applicable for in-line processes. Here, the development of an infrared (IR) thermo-graphic imaging diagnostic for gas diffusion electrodes (GDEs) is reported on. This technique employs a reactive-flow-through (RFT) excitation by reacting a stoichiometric, non-flammable H₂/O₂ gas mixture in balance N₂ on the catalyst of GDEs. The resulting heat signature contains information about the Pt content and its variations. This heat signature is measured using an IR sensor. The concept was selected because it was promising to be feasible for an ambient manufacturing environment.

The use of RFT is attractive for the evaluation of the catalyst layers on GDEs due to ease of implementation and low cost of gas delivery systems. The technique complements a previously developed in-situ technique for catalyst-coated membranes (CCMs) that employed an electrical in-plane current through the electrode layer to create a heat signature [3]. This method was not applicable for gas diffusion electrodes because the highly conductive carbon material of the gas diffusion layer (GDL) prevented electrical excitation of the electrode layer. The RFT technique is an alternative excitation method that employs a catalytic exothermic reaction to create the heat signature. The work here is a continuation of initial studies that focused on the proof of principle
of RFT. The development of adapting RFT to an ambient environment with moving samples and an impinged flow of the reactive gas, i.e. a mass production environment is reported here.

1.1. **Introduction of PEMFCs and the motivation for research**

The first application of the PEMFC was as an auxiliary power source in the 1960s for the Gemini space flights. Original PEMFCs featured catalyst loadings upward of 4 mg cm$^{-2}$. With such high Pt loadings, the cost of platinum was the main reason fuel cells were not commercialized [18]. Today, the membrane electrode assembly (MEA) is still a major obstacle for commercialization of PEMFCs because the cost of electro-catalysts is a significant cost factor for material costs [2]. Reducing the catalyst loading and improving catalyst utilization is pinnacle for improving the commercial viability of fuel cells [16]. The primary motivation behind developing quality control techniques is ultimately to reduce the cost of manufacturing diagnostics used on fuel cells and to improve the life span of fuel cells by ensuring that manufactured components are produced without defects.

1.2. **PEMFC functionality**

A polymer electrolyte membrane fuel cell (PEMFC), as state above, is an electrochemical energy conversion device that converts hydrogen and oxygen (or air) into heat, electricity, and water. A PEMFC consists of a membrane electrode assembly (MEA), a bipolar plate (BP), humidifiers, blowers, and valves. The MEA consists of proton diffusive membrane, an anode, and a cathode. At the anode side of a PEMFC, platinum catalyst splits hydrogen into protons and electrons forcing the electrons to pass through the bipolar plate to the cathode side of the fuel cell. Protons are diffused through the ionomer membrane separating the electrodes. At the cathode side of the fuel cell, oxygen is split at the platinum catalyst and combined with the protons and electrons to form heat and water. A schematic for a PEMFC is provided in Figure 1.1. PEMFCs are often combined in series to form a fuel cell stack to generate high voltages and power from the combined cells for effective use in daily application. The fuel cell stack may be connected in parallel if the stack is not large, with each cell is separated by a bipolar layer [15, 19, 32, 33].

1.3. **Applications of PEMFCs**

The primary areas of interest for using PEMFCs are in transportation and cogeneration with methanol-reformed fuel. Mass transit buses implementing PEMFCs have been developed by
Ballard Power Systems with the latest power rating exceeding 300 kW. In this particular application, hydrogen is typically stored in compressed gas cans made of fiberglass-supported aluminum. Fuel cells have also been implemented in smaller vehicles. Other examples of applications include 10 kW-rated PEMFC power generators operating on hydrogen supplied from a methanol-reforming process [1, 15, 33].

Figure 1.1: Schematic for a polymer electrolyte membrane fuel cell

1.4. MEA components

The MEA consists of an anode, cathode, gas diffusion layer, and proton diffusive membrane. The anode and cathode are typically either a gas diffusion electrode (GDE) or a catalyst-coated membrane (CCM). GDEs are composed of a gas diffusive layer (GDL), a platinum/carbon catalyst layer (CL), and a micro-porous layer between the GDL and the CL. A CCM consists of the proton diffusive membrane with the anode on one side and the cathode on the other. The catalyst layer for PEMFCs typically contains Pt on a carbon support mixed with
an ionomer electrolyte [21, 33]. The ionomer allows for proton transfer within the anode, cathode, and the membrane [5, 33]. Catalyst layers are generally heterogeneous and have thicknesses between 5-50 µm [24-26, 31, 33]. Optimizing the catalyst layer loading is one of the key points of research for reducing the cost of MEAs. The latest research in PEMFCs seeks to reduce the catalyst loading to below 0.4 mg Pt cm⁻² [8, 18].

The gas diffusion layer (GDL) is a heterogeneous porous carbon paper or cloth mixed with polytetrafluoroethylene (PTFE) with thickness between 100-300 µm that allows for diffusion of gas, electric current, and heat produced at the catalyst layer [18-19, 21, 23, 26]. The GDL is positioned between the CL and the bipolar plate. The amount of PTFE is generally between the range of 15% and 40% by weight [33]. Gas diffusion layers are designed to balance removal of excess water while simultaneously diffusing reactant gases to the catalyst layer and allowing enough hydration to promote proton conductivity [18-19, 23]. GDLs are generally coated with PTFE for its hydrophobic properties [18-19, 26].

The micro-porous layer is a thin carbon agglomerate layer added between the catalyst layer and the gas diffusion layer in order to provide a more uniform interface between the GDL and the CL. MPLs reduce effects of flooding in the GDL and allow for better gas transport. MPLs can also resist compression of the CL, which can cause embedding into the GDL, reducing activity [32].

The proton exchange membrane is approximately a 1 mm thick ionomer layer sandwiched in between the anode and cathode. The purpose of the proton exchange membrane is to transport protons produced at the anode to the cathode catalyst layer where they combine with electrons and oxygen to form water and produce heat [32, 33].

GDEs consist of the gas diffusion layer with a micro-porous layer and an electrode. According to Rajalakshmi et al, GDEs require the following qualities to be effect and viable: (1) a large interface between the polymer electrolyte and the catalyst layer, (2) effective transport of protons from the anode to the cathode, (3) effective transport of gases and removal of condensed water, and (4) effective current collection [27]. The most common electrode design is a thin-film design, which is characterized by a thin Nafion film that binds the carbon-support catalyst particles. The thin-film design, however, has reduced resiliency [18].
1.5. Fuel Cell Manufacturing and Production

Several groups have done research on cost analyses for fuel cells. Tsuchiya et al explored economically, the viability of fuel cells given development of mass production techniques and a learning curve for lowering costs of manufacturing [32]. Currently, the cost of Pt is $5 / kW [32]. According to Tsuchiya et al, the goal of the automobile industry is to reduce the cost of PEMFC stacks to $40/kW [32]. Bar-on concluded that approximately 500,000 fuel cell units per year must be produced using mass production facilities in order for PEMFCs to be viable based on two different cost analysis studies [8].

MEAs are assembled by hot pressing the proton exchange membrane between the anode and the cathode. CCMs and GDEs are manufactured in roll-to-roll processes where the coatings are applied to the roll and the finished product is wound into a final roll [3,11]. A method of manufacturing GDEs by mixing catalyst and ionomer while simultaneously pouring onto an electrode belt has been explored [10]. The carbon used for the GDL is generally manufactured in four steps: (1) pre-pegging (strands are aligned with spools followed by a resin bath), (2) molding, (3) carbonization, and (4) graphitization [18]. Catalyst layers are generally manufactured during assembly of the MEA, while the membrane and GDL are manufactured prior to assembly. Preparation of catalyst layers is a vital aspect for reducing costs of GDEs [20]. Catalyst layers can be manufactured a number of ways such as screen-printing, brushing, and electro-spraying. Screen-printing is one of the most effective means of manufacturing the catalyst layers for GDEs [27]. The electrospray technique is much more effective than brush techniques [26].

Fuel cell performance is dependent on a number of component parameters, such as catalyst loading of the anode and cathode, the thickness of the CL and GDL, porosity of the GDL and CL, etc. [19]. Xianguo et al compiled a list of parameters for each component that have an effect on the fuel cell performance [33]. Much research is conducted on studying the effects of changing a parameter. As an example, the effect of PTFE in fuel cell has been studied extensively. Giorgi et al conducted research on the impact of PTFE on the performance of a fuel cell and discovered that increased PTFE led to a greater amount of oxygen at the catalyst sites as well as a decrease in porosity of the GDL. He concluded that minimizing PTFE without promoting water flooding and keeping the GDL structurally sound was optimal [12]. Research
has been conducted on the effects of variations in GDLs [16]. The effects of reduced platinum yields a drop in performance of the fuel cell [2]. Knowing this, it is important to reduce the presence of defects in fuel cell stacks to increase reliability and to maintain optimal performance.

1.6. **Defects in Gas Diffusion Electrodes**

In addition to reducing the cost of materials in the manufacturing process, detecting defects in fabricated membranes is pertinent to ensuring that produced material is reliable [6]. Defects can lead to loss in performance and/or failure in the fuel cell stack at the defect area [3,11]. One reason for performance loss is an uneven compression of the GDL at the location of the defect, which often results in flooding or rupture [3,11]. Ahn et al conducted operation of a 40 cell PEMFC with a maximum performance of 2.89 kW with pure oxygen, but operation of the PEMFC showed rapidly declining performance of 1800 hours. The result of failure was catalyst degradation and contamination of the PEM [1]. It is likely that a decrease in compression can be caused by a defect with reduced catalyst. An agglomeration of catalyst would thus yield an increase in compression [28].

1.6.1. **Definition of a defect**

A defect is a non-uniformity in the thickness or loading of a catalyst layer such as a scratch, abrasion, agglomeration of catalyst, crack, void, pinhole, or thickness variation that can cause a PEMFC to fail because of pressure variation or early rupture of the MEA [3]. A defect can be a decrease or an agglomeration of catalyst. Defects can come in all shapes and sizes, but as expected, the larger and more severe the defect, the greater chance for failure to occur. Defects not detected before implementation of the GDE in PEMFCs can lead to failure of fuel cells, which is not only expensive, but also time intensive because of the amount of time it takes to salvage material and rebuild the fuel cell. Defect detection that is fast, non-destructive, and relatively inexpensive is necessary for the mass production process and is the object of this project [11]. Defect modeling using COMSOL has been conducted by various groups such as Das et al [11]. Hinaje et al also generated a 2D COMSOL model of a defective GDL sample for a pinhole defect and water droplet and yielded results that show a drop in voltage with the defects [14].
1.6.2. **Defect Detection**

Detecting defects is a major milestone in the manufacturing process of MEAs and GDEs. The ability to inspect new material for defects is key for reducing failure in fuel cell stacks and ensuring that produced material is pristine [3,11]. Most techniques for detecting defects have been classified into one of two categories: Electrochemical tests, which test electrical properties, such as Electrochemical Impedance Spectroscopy (EIS), and physical tests that measure physical properties such as pressure [7]. Visual inspection and point inspection of defects are the most natural methods of detecting defects, but are not advantageous for MEAs because (1) defects are not always visible on the black GDE surface and (2) point measurements only provide statistical data and cannot detect discrete spot defects [3]. X-ray Fluorescence (XRF) has been proposed as a viable detection technique, but XRF is not feasible for scanning large areas [11]. IR thermography cannot identify element specific information regarding the catalyst loading, but can map catalyst thicknesses [11].

1.7. **Infrared (IR) Thermography**

Infrared (IR) thermography is the method of creating images that correspond to the temperature profile of an object based on emitted blackbody radiation in the infrared wavelength spectrum. IR thermography is based on the idea that objects emit radiation in well-defined bands of electromagnetic frequencies based on the temperature of the object. Generally speaking, the hotter the object is, the higher the frequencies of emitted radiation. A perfectly black body will emit Blackbody radiation, but we can relate the power of the emitted radiation for any object to its surface temperature using the Stefan-Boltzmann relation:

\[
E = \epsilon \sigma A T_s^4
\]

where \( E \) is the emitted radiation in W/m^2, \( \epsilon \) is the emissivity of the surface, \( \sigma \) is the Stephan-Boltzmann constant equal to 5.67 X 10^8 W m^{-2} K^{-4}, \( A \) is the surface area, and \( T_s \) is the surface temperature in Kelvin [9].

Objects near room temperature emit radiation in the infrared spectrum, with wavelength ranges roughly between 1 µm – 100 µm, and peak radiation near 10 µm [9]. The IR camera used in this work was a Jenoptik Vario-Cam HiRes camera with a 640 by 480 pixel detector with a temperature accuracy of ±1.5K. The spectral bandwidth of the camera was 7.5 µm – 14 µm, making it sensitive enough for room temperature measurements. The camera in conjunction with
IR cameras’ Thermography Suite software allowed for video and single image capture, as well as data extraction and analysis.

IR thermography is useful for characterizing surface properties that cannot be mapped using visible spectrum imaging. IR imaging has been used for studying properties in sheet metal production [4], building diagnostics [7, 20], medical research [29-30], characterizing cracks and folds in membrane materials [13], and for characterizing PEMFC parameters [11]. Previous work using IR thermography for fuel cell diagnostics includes work by Yamada who explored IR thermography to evaluate anode catalyst layers in PEMFCs [33], Aieta et al who explored IR thermography with DC excitation to study variations in catalyst thickness [3], and Das et al who explored IR thermography with reactive-flow-through excitation to detect defects with stationary environment [11]. These researchers demonstrated that heterogeneities, whether defects or thickness variations in fuel cell materials, may not be visible in the visible light spectrum, but may be visible when the material is thermally excited and viewed with an IR camera.

1.8. Reaction Flow-through Excitation

An exothermic gas reaction between hydrogen and oxygen can be induced at a platinum catalyst even if the concentration of hydrogen is less than the flammability limit of 4% H₂ in air or stoichiometric oxygen. By impinging a gas flow onto the catalyst layer of a GDE, it is possible to create a detectable heat signature with an IR sensor. Thermal excitation is very strong with even low hydrogen concentrations (0.4%-2.0%). Das et al determined that the strength of the thermal response was directly dependent on the defect loading and gas flow-rate and had little to do with the size of the defect. They also noted that the hydrogen concentration had a greater effect on the thermal response than the flow-rate because of convective cooling. Further, the hydrogen gas-reaction technique was determined to not have any corrosive properties on the carbon support or result in performance loss [11]. As mentioned above, the technique of delivering a hydrogen and oxygen gas mixture to a GDE catalyst layer has been name reaction flow-through (RFT).

1.9. Objectives

The main objective of this project was to detect defects on gas diffusion electrodes using IR thermography with a reaction flow-through excitation using a stoichiometric gas mixture of H₂ and O₂. Defects with at least a 1°C difference from the surrounding temperature profile were
considered detected in the previous IR DC excitation study [3]. A 5mm by 5mm defect and 100% reduction in catalyst was set as the project’s benchmark case. As there is no industrial standard for common defect sizes, this size defect was elected to be representative of a common defect size. A 2 mm by 2 mm defect was detected in order to exceed this baseline case. After the proof of concept of the RFT technique, the long-term goal of the research is to implement the diagnostic on a web-line similar to the one shown in Figure 1.2. The following chapters explain the progress made towards meeting this objective and explain the natural progression from a stationary, closed environment to an open, moving environment.
The purpose of the stationary experiments was to characterize the thermal response of the GDE with RFT excitation and to get a preliminary understanding of the potential to detect defects. The stationary experiments proceeded as follows: (1) defects on a GDE sample were detected in a brass enclosure as shown in Figure 2.1, (2) a pseudo gas knife consisting of a piece of gasket material with a slit (slit width of 1.81 mm) was inserted into the closed assembly to simulate an gas knife gas stream on the material, and (3) an gas knife was created and used on a stationary piece of GDE to test the thermal response in an open environment. The use of brass as the manifold material was arbitrarily chosen.

2.1. Closed environment with brass manifold experiments

An image and exploded view diagram are provided in Figure 2.1. The purpose of the closed experiments was to demonstrate that RFT could produce a thermal response strong enough to detect heterogeneities in the material. If the closed environment experiments produced promising results, then the techniques would be applied to an open environment. The closed environment testing utilized a brass manifold top and bottom with polymer gasket seals and screw fasteners. A flow stream could be initiated at the top or bottom of the manifold with the outgoing flow going to an exhaust vent. Gas flow was controlled by a 5000 standard cubic centimeter per minute (SCCM) maximum flow controller made by MKS (model M100B01353CR1BV). The top of the manifold contained an infrared transparent film to permit imaging of the GDE inside the manifold.

The first set of experiments with the brass manifold was conducted to determine if the thermal response and detection capabilities of the RFT and IR thermography technique following excitation were adequate. These tests involved a series of different platinum-loaded GDEs with a variety of defect sizes and catalyst reduction percentages. Provided in Figure 2.2 is a sample IR image frame at thermal steady state (point where temperature stops changing) at a flow-rate of 5000 SCCM with 0.4% H₂ concentration.

As can be seen from the IR image (Figure 2.2), the reaction between hydrogen and oxygen gas was significant enough to induce a noticeable increase in temperature as detected by
the IR camera. A 1 cm² square defect with 25% less catalyst is applied on the center of the GDE. The reduced catalyst should correspond to a weaker thermal response. The 1 cm² is identifiable in the center of the GDE on the false-image plot as having a reduced temperature (Figure 2.2). In order to get a better idea of how well the defect is actually being detected, a temperature analysis line was drawn through the middle of the defect. A plot of the temperature vs. the line length is provided in Figure 2.3.

Figure 2.1: Closed experiment brass manifold test chamber (left) and an exploded view of brass manifold with pseudo-gas knife gasket insert or slit gasket (right). The area of active excitation is 4.5 cm x 4.5 cm.

The line analysis reveals that the defect is noticeable at pixel 300 with a 0.5°C decrease from the left peak and a 1°C decrease from the right peak. Given a 1°C decrease criteria, the defect is detectable. Given that the defect’s catalyst reduction is only 25%, the IR thermographic technique can detect defects that still have a significant catalyst loading (~75% of original catalyst loading).
2.1.1. **Pressure hysteresis test**

To determine if the porosity of a GDE significantly changes after prolonged exposure to gas flow, a pressure hysteresis test was performed. A change in porosity indicates that the GDE is physically altered as a result of the gas flow and thus the RFT excitation technique is damaging GDE. This experiment was conducted by flowing gas into a brass manifold with a pressure transducer (operating pressure range of 0 psig to 7.1 psig, Omega HHP352b) connected at the top and bottom of the manifold. The pressure transducer measures the pressure drop as a result of gas flow passing through the GDE. A baseline gas flow-rate was used prior to running the test flow-rates. After the test flow-rate was ran, the baseline flow-rate was ran again. The starting baseline flow-rate pressure differences measured by the pressure transducer were compared with the ending baseline flow-rate pressure differences. The first two test flow-rates were 200 SCCM and 500 SCCM with a baseline flow-rate of 100 SCCM. All following tests had flow-rates of 1000 SCCM through 5000 SCCM with 500 SCCM steps and a baseline flow-rate of 500 SCCM. Provided is a table (Table 1) with the pressure differences for the initial flow-rate pressure differences, test flow-rate pressure differences, and the final flow-rate pressure differences.

![Figure 2.2](image.png)

Figure 2.2: IR image of steady state temperature response for a defective GDE sample with 0.2 mg Pt cm\(^{-2}\) and 1 cm\(^2\) defect with 25% reduction in loading in a brass manifold with 0.4% H\(_2\) concentration and 5000 SCCM flow-rate

The pressure did not change even at the highest flow-rate before and after applying the test flow-rate. This indicates that the GDE is not physically altered as a result of the gas flow.
Since the porosity has direct effects on the resulting pressure difference at a given flow-rate, a change in pressure before and after the high gas flow would imply material alteration. Overall, the experimental results indicate that the gas flow is not altering the material in a significant manner.

![Temperature vs. Length for Closed Manifold Experiment](image)

Figure 2.3: Line temperature vs. length analysis of sample steady state for closed manifold experiment through the center of the defective area on the defect

<table>
<thead>
<tr>
<th>Baseline Flow-rate (SCCM)</th>
<th>Pressure (psig)</th>
<th>Test Flow-rate (SCCM)</th>
<th>Pressure (psig)</th>
<th>Final Flow-rate (SCCM)</th>
<th>Pressure (psig)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.0055</td>
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### 2.2. Pseudo-gas knife experiments

The purpose of the pseudo-gas knife experiments was to simulate gas delivery in an open environment by creating a knife-like flow similar to what would be used in a manufacturing environment, while retaining some of the infrastructure of the manifold experiments. These experiments were used to determine how the open environment affects the quality of thermal response after RFT excitation. The pseudo-gas knife experiments were also used to detect defects in GDE materials. These experiments utilized the brass manifold and the pseudo-gas knife for gas delivery to the substrate, but the GDE was suspended in the outside air instead of being contained within the manifold.

#### 2.2.1. Loose GDE experiment

![Figure 2.4: Loose gas diffusion electrode experimental setup](image)

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</table>
The purpose of the loose GDE experiment was to primarily test the effects of gas delivery from the gasket gas knife in an open environment while simultaneous testing stationary defect detection on the GDE. The manifold top piece was removed, exposing the GDE to the open air. The experiment involved laying a piece of GDE on rubber gasket pieces (2 cm thick) over the pseudo-gas knife opening while running gas flow onto the GDE (See Figure 2.4 for an image of the setup). The impinging gas flow had a stoichiometric H₂/O₂ with a 0.4% H₂ concentration. The desire was to generate a relatively fast response time that in turn would give an idea of how viable a moving substrate environment would be. Flow-rates of 500 SCCM to 5000 SCCM in 500 SCCM increments were tested. A set of steady state temperature IR images for flow-rates of 1500, 2000, 2500, and 3000 SCCM is provided in Figure 2.5.

Figure 2.5: Loose GDE experiment steady-state IR frames of a defective GDE with 0.2 mg Pt cm⁻² loading and a 2 cm² square defect with no catalyst present for flow-rates: 1500 SCCM (top left), 3000 SCCM (top right), 4000 SCCM (bottom left), and 5000 SCCM (bottom right).

The loose GDE experiment image frames show a thermal response similar to IR analyses conducted on the closed environment experiments. The defect is clearly visible in the center of
the IR image. As the flow-rate was increased, the thermal response surrounding the defects increased. The excitation did not conform to the shape of the slit in the pseudo-gas knife. This is likely a result of the sheet of gas being distorted by the environment. The reason for more excitation of the GDE on the right side of the frame is unknown. In order to gain a quantitative understanding of how detectable our defects are, a line analysis was conducted for each case in Figure 2.6.

![Figure 2.6: Line temperature vs. length analysis through center of defect for the loose GDE experiment for flow-rates: 1500 SCCM, 3000 SCCM, 4000 SCCM, and 5000 SCCM.](image)

The line analyses reveal a drop in temperature where the defect exists. The edges of the defect correspond to the sharpest slope in the line analysis. As the flow-rates were increased, the temperature surrounding the defect increased, while the temperature at the defect increased only slightly. This slight increase in temperature is likely a result of heat conduction. Overall, it was determined that the effects of the open environment were not significantly affecting thermal response or the ability to detect defects.

The purpose of the suspended GDE experiment was to test the effect of the height between the pseudo-gas knife and the GDE with the catalyst facing down. This information is useful for
understanding an acceptable range of heights between the GDE surface and the gas source when conducting moving experiments. The setup height of the GDE was controlled using screw bolts and zip-ties. The height was set with a ruler measurement. The impinging flow consisted of a stoichiometric non-flammable 0.4% H₂/O₂ in balance N₂ gas mixture flowed at 5000 SCCM. A picture of the setup is included in Figure 2.7. A set of sample IR images at heights: 1.5 cm, 2.0 cm, 2.5 cm, and 3.0 cm for a defect-free GDE with 0.2 mg Pt cm⁻² is provided in Figure 2.8.

2.2.2. Suspended GDE experiment

As seen in the sample IR images, the response yielded a noticeable thermal response for heights 1.5 cm and 2.0 cm, but the excitation was not uniform or symmetric. The thermal responses for the 2.5 cm and the 3.0 cm cases were poor. As with the loose GDE experiment, we do not see a slit-like image despite the inclusion of the pseudo-gas knife. A line analysis was conducted to generate a temperature profile for each of the heights, and the plot is provided in Figure 2.9.
From the above plot, we see that at our maximum temperatures for the 1.5 cm height case was approximately 28.6°C. This is a temperature increase of approximately 3°C from the surrounding temperature. As expected, the closer the gas source is to the GDE surface, the stronger the thermal response will be. While the thermal response yielded a large increase in temperature at 1.5 cm, it is likely that the height will need to be much smaller to accommodate a moving substrate. Lower heights were not tested because it was not expected that the operational height between the gas knife and substrate would need to be only a few mm. Experimental results yield a non-uniform temperature profile. The cause of the non-uniformity is uncertain, but it may be that the gas flow out of the slit is coming out at an angle because gas flows into the lower half of the manifold from the side. If the resulting gas flow is not directly flowing upward, then a thermal response will be seen on one side of the GDE as opposed to the middle. This experiment should be conducted again in order to test heights that are more applicable in actually operation and with a more uniform gas flow from the slit gasket.

![Figure 2.8: Suspended defect-free GDE with 0.2 mg Pt cm⁻² loading experiment steady-state IR frame for heights: 1.5 cm (top left), 2.0 cm (top right), 2.5 cm (bottom left), and 3.0 cm (bottom right)](image)
2.3. **Perforated tube gas knife**

In order for a successful transition to the moving environment, being able to deliver the gas mixture to the substrate surface quickly was pertinent. The gas delivery must be delivered in a sheet-like gas flow as the GDE material passes through the manufacturing web-line. This prompted an in-house design of a perforated tube gas knife. The designed gas knife is a 1/2” OD with a 3/8” ID stainless steel tube with 115 0.5 mm diameter drilled holes symmetrically along the tube with 2 mm between each hole. At each end of the gas knife is a 1/4” Swagelok male connection. Diagrams of the entire knife and a close up of the holes are provided in Figure 2.10 and Figure 2.11 respectively. Gas knife characterization is covered below.

![Temperature vs. Position for Suspended GDE Experiment](image)

**Figure 2.9:** Line temperature vs. length analysis of suspended GDE experiment plot for heights: 1.5 cm, 2.0 cm, 2.5 cm, and 3.0 cm

![Perforated tube gas knife](image)

**Figure 2.10:** Perforated tube gas knife.
2.3.1. **Gas knife holder**

In order to hold the gas knife in a manner that allows for reproducible and precise experiments, an gas knife holder was designed. The gas knife holder consisted of a rod and housing section with pinholes for height setting and a cradle style clamp that allowed for angled adjustment. The housing and rod both were inscribed with tick marks to mark the height of the knife relative to the GDE material. The cradle clamp was inscribed with angle demarcations in order to set the gas knife angle. The height adjustment allowed for a 1 mm incremental change in height with a range of 10 mm above and below the GDE surface. Pictures of the gas knife holder are provided in Figure 2.12.

![Perforated tube gas knife (Close-up)](image)

Figure 2.11: Perforated tube gas knife (Close-up)

2.3.2. **Gas knife characterization**

Before conducting reaction flow through testing with the gas knife, a series of characterization experiments were performed in order to access the performance and uniformity of the perforated tube gas knife. Tests included hotplate tests to measure convective cooling effect and uniformity, angled gas knife tests to measure flow fluctuations due to the finite separation of the gas knife holes, and microscope imaging to access the quality of the gas knife machining.
In order for a thorough analysis, sample IR images were collected at the steady state of each respective case. A line analysis was conducted on the areas near the gas knife as a temperature vs. distance plot. For the hot plate experiment, the analysis of the area directly underneath the convective cooling area was subtracted from a line analysis offset to a part of the hot plate that was not being cooled by the knife. Because the temperature profile was round, the resulting subtraction led to a sharp slope at the edges. The sample IR image and the line analysis plots for the hot plate experiment are provided in Figure 2.14 and Figure 2.15 respectively.

![Gas knife holder (assembled and separated)](image)

2.3.3. **Hot plate uniformity experiment**

The purpose of the hot experiment was to image the uniformity of the gas knife by impinging flow onto a laboratory hot plate. The gas knife was setup directly over a standard laboratory hot plate with the impinging gas flow set 45° to the hot plate surface and a height above the hot plate of approximately 2 mm. The following conditions were test: flow-rates of 5000 SCCM and 20,000 SCCM, hot plate temperatures of 40°C, 60°C, 80°C, and 100°C, and gas flow from the left, right, and both sides of the gas knife. A picture of the setup is provided in Figure 2.13.

As we see from the IR frame and the plot, the effects of convective cooling are significant. The cooling is visually detectable, and a line analysis shows a significant drop in temperature of approximately 12°C. The noise from the hot plate analysis line suggests that the
gas knife was non-uniform. These experiments should be conducted again with variable heights to understand how the uniformity of the convective cooling changes with changing the height between the hot plate and gas knife.

Figure 2.13: Hot plate experimental setup

Figure 2.14: Sample IR image of steady state convective cooling of a hot plate at 100 degrees Celsius by an gas knife
Figure 2.15: Line temperature vs. length analysis of convective cooling pattern for the steady state hot plate experiment

Figure 2.16: Bench-top Roller schematic with angled camera and orthogonal camera orientations

A schematic for the bench-top roller setup is provided in Figure 2.16 and a picture of the bench-top roller system configured for the angle experiment is provided in Figure 2.17. The
bench-top roller was designed by the fuel cell manufacturing group at NREL. The purpose of this experiment was to visualize the thermal response directly underneath the gas knife to understand how the impinging gas flow was affecting the GDE surface and the catalytic reaction. The gas knife is directly above the area of excitation; and therefore, an angled camera is required to image the excitation at impingement. The GDE material was held stationary while being thermally excited to steady state.

The angled camera experiment was conducted on a bench-top roller system designed by the NREL fuel cell manufacturing group and consisted of two sets of brass rollers with one set controlled by an electric motor. The GDE had a nominal catalyst loading of 0.4 mg pt cm\(^{-2}\) and was defect-free. The GDE was held between a pair of the brass rollers while one of ends hung freely. The material was oriented with the catalyst loading facing upwards towards the gas knife, while the gas knife holes faced downwards towards the GDE material. The camera was set at a 60° angle relative to the surface of the GDE as shown in Figure 2.17. The height between the GDE and the gas knife holes was set to 1 mm. A 2% H\(_2\) / 1% O\(_2\) in balance N\(_2\) gas mixture was flowed onto the GDE surface at a flow-rate of 20 standard liters per minute (SLPM) through the gas knife.

2.3.3.1. Results

The RFT excitation generated a spatially oscillating thermal response that matched closely the spacing of the gas knife holes. Convective cooling as a result of the impinging jets likely causes this. The IR image is provided in Figure 2.18. The excitation area was then analyzed with a series of temperature lines placed at increasing offset distances from the gas knife. The first line was centered at peak excitation with the second line adjacent to the first. The third line was placed 2 mm from the second line, and fourth line was placed 2 mm from the third line (See Figure 2.14 for clarity). These auxiliary lines were included to study the conduction through the material outward from the excitation spot. By comparing line one and two, we can understand the effect of convective cooling relative to RFT excitation. A line analysis temperature vs. distance plot is provided in Figure 2.15.
2.3.3.2. **Bench-top Roller setup for angle experiment**

![Angular setup image](image)

Figure 2.17: Angled camera experimental setup

The resulting profile for analysis line 1 is oscillatory as a result of the cooling spots. The cooling spots are located at the troughs along the line. Because convective cooling is most apparent at the point of impact, the surround temperatures are much higher than the hole temperatures. These higher surrounding temperatures are inducted on the line as the crests. The analysis line 2 temperature vs. distance profile conforms to the average shape of analysis line 1. This implies that the thermal response along analysis line one is approximately equivalent to the thermal response along analysis line 2. Analysis lines 3 and 4 show a decrease in temperature in accordance to conduction and convective cooling to the environment. Overall, effects of convective cooling from the individual holes are significant as demonstrated by analysis line 1.

2.4. **Summary**

The transition from closed environment IR thermography with RFT excitation to an open environment and the development of the gas knife was conducted. The open environment did not significantly alter the ability to detect a defect on a GDE. The height relationship between a GDE
surface and a gas source and the thermal response was characterized. An gas knife was designed and characterized for uniformity using thermal response profiles.

Figure 2.18: Thermal image frame of stationary angled camera experiment showing thermal excitation due to jet impingement

Figure 2.19: Temperature vs. position line analysis directly over holes (analysis line 1), directly adjacent to holes (analysis line 2), 2 mm offset distance from analysis line 2 (analysis line 3), and 2 mm offset distance from analysis line 3 (analysis line 4).
CHAPTER 3 MOVING GAS DIFFUSION ELECTRODE EXPERIMENTATION

With a perspective of the stationary thermal response behavior of the GDE and the RFT excitation, a moving environment was tested next. A moving environment presented different challenges such as convection caused by the moving substrate, reduced gas/catalyst contact times, and uncertain gas delivery to the substrate. This chapter focuses on characterizing the thermal response of a moving GDE by changing the gas concentration, gas flow-rate, gas knife height, gas knife angle, gas knife and camera orientation, and GDE speed. By understanding the transient thermal response, defect detection becomes easier to predict. The purpose of the moving substrate experiments was to characterize the diagnostic under conditions typical of a manufacturing environment. Understanding how the material behaves under different conditions was vital to understanding the potential for the RFT diagnostic.

3.1. GDE material preparation

An example picture of GDE material with leaders and trailers is provide in Figure 3.1. The GDE material was cut into samples suitable for the moving test environment. The GDE material was provided by Johnson-Matthey as sample sheets with nominal platinum catalyst loadings of 0.4 mg Pt cm\(^{-2}\). Due to the limited size of the GDE samples, a leader and trailer sheet of GDL material were attached to the GDE material to maintain tension during experimentation. The GDL material also allowed for a pre-experiment gas flow in order for excitation to occur as soon as the GDE passes underneath the gas source, allowing us to image the entire GDE material.

3.2. Bench-top roller setup for transient experiments

The bench-top roller system was utilized to conduct transient experiments on the GDEs. The bench-top roller system consisted of two sets of brass rollers with one set powered by a speed adjustable electric motor. As the GDE material is propelled in between the brass rollers, the material passes underneath the gas knife with the catalyst layer facing upwards towards the holes facing downwards. The gas knife holder allowed for height and angle adjustment of the gas knife in order to test the sensitivity of the GDE to variations in height and angle. The IR camera is place directly above the location where thermally excited GDE comes into view.
3.3. **Experiments**

The following experiments sought to characterize the RFT technique and its ability to thermally excite the moving GDE while imaging the thermal profile using an IR camera. The first set of experiments sought to alter parameters (flow-rate, gas concentration, height, and angle) while holding all other conditions constant. The second set of experiments was intended to characterize the effect of substrate speed. The last set of experiments sought to explore the geometric effects of the gas knife for uniformity of the thermal response when flow-rates were varied by taping over the gas knife holes to scale up to a higher flow-rate.

![Image 1](image1.png)

**Figure 3.1:** Gas diffusion electrode (center) with taped on gas diffusion layer leader and trailer.

3.3.1. **Thermal response vs. flow-rate and H\textsubscript{2}/O\textsubscript{2} concentration**

The purpose of this experiment was to study the thermal response of the moving GDE as the flow-rate and the H\textsubscript{2}/O\textsubscript{2} concentration were varied. Understanding the thermal response behavior of the GDE with respect to parameters gives insight to the best conditions for exciting the GDE with as low of a flow-rate and hydrogen concentration as possible and still detects defects. A set of temperature vs. time collection points was drawn parallel to the gas knife at the point of the maximum excitation (See Figure 3.3 for an example). These temperature collection points contain temperature vs. time data. These collection point temperatures were averaged at each time step (spatial averaging) plotted. Flow-rates of 1500 SCCM, 3000 SCCM, and 5000
SCCM were tested for concentrations of 0.4% and 2.0% respectively. The flow rate per distance of gas knife holes was also included in the legends for thoroughness. The GDE was moving at a velocity of 10 feet per minute (fpm). The moving average probe point temperature vs. time plots for the flow-rates and concentrations of 0.4% and 2.0% H₂/O₂ are provided in Figure 3.4 and Figure 3.5.

![Bench-top roller setup](image)

Figure 3.2: Bench-top roller setup

As seen in the temperature vs. time plot (Figure 3.4) for the 0.4% H₂ case, increasing the flow-rate has a very limited effect on the thermal response of the GDE. The gas concentration of H₂ is very low and as a result, even high flow-rates do not yield a strong thermal response. At the 5000 SCCM case, only a 1°C is seen with practically no increase in temperature for the 1500 SCCM and 3000 SCCM case. It is important to understand how increasing the hydrogen concentration will affect the relationship between the thermal response and the flow-rate.

The 2.0% concentration case yielded a stronger thermal response. We see approximately a 10°C increase in temperature at 5000 SCCM, approximately 6°C increase in temperature at 3000 SCCM, and approximately a 3°C increase for 1500 SCCM. The result of using 5x more hydrogen and oxygen is demonstrated by a greater extent of reaction and a higher temperature
profile. These results suggest that the concentration of hydrogen and oxygen is more influential on the resulting thermal response than increasing the flow-rate.

Figure 3.3: Example IR image with temperature collection points

3.3.2. **Thermal response vs. gas knife offset height**

The purpose of the height experiments was to understand how much gas knife height above the GDE influenced the thermal response on a GDE with all else constant. The heights compared were 1 mm, 2 mm, 3 mm, 4 mm, and 5 mm. The gas knife angle was kept orthogonal to the GDE surface. The flow-rate was kept at 2% H₂ and 5000 SCCM, and the GDE was moving at a velocity of 10 fps. The temperature vs. time data points were collected and analyzed similarly to the thermal response vs. flow-rate and concentration experiment. The temperature data points were plotted in Figure 3.6.

The temperature vs. time plot shows exactly what is predicted as the height is varied. The temperature after excitation decreased as the height increased. This makes sense because if the
height between the gas source and the substrate is large, more of the gas will escape into the environment. Less excitation yields a lower temperature on the GDE. According the plot, the 1 mm case yielded a temperature increase of approximately 10°C. The 2 mm case yielded a temperature increase of approximately 8°C. The 3 mm case yielded a temperature increase of approximately 6°C. The 4 mm case yielded a temperature increase of approximately 3°C. Lastly, the 5 mm case yielded a temperature increase of approximately 1°C. These temperature increases indicated a strong relationship between thermal response and height, as expected. It is important to note that time dependent drops in temperature occur at approximately 0.4-0.5 seconds. The reason for this predictable drop is that the tape holding together the GDE and the GDL creates a vibration as it passes through the brass rollers. Because the thermal excitation of the GDE is sensitive to any height changes, the vibration will create a noticeable drop in temperature as the tape passes through the rollers. When the vibration stabilizes, the thermal response returns to normal. This explains the drop followed by the rise.

Figure 3.4: Thermal response vs. time with 0.4% H₂

3.3.3. **Thermal response vs. flow angle**

The purpose of this experiment was to test how the angle of the gas flow from the gas knife affected the thermal response. The angles compared were 30°, 45°, 60°, and 90° where 90° is a gas flow orthogonal to the surface. The gas flow-rate was 500 SCCM, the gas composition
was a stoichiometric non-flammable 2% H₂/ 1% O₂ in balance N₂ gas mixture, and the GDE was moving at a velocity of 10 fpm. The average temperature vs. time data points were analyzed and are plotted in Figure 3.7.

Figure 3.5: Thermal response vs. time with 2.0% H₂

As expected, the thermal response decreases as the angle of the gas knife gas-flow becomes less orthogonal to the substrate. This makes sense because the less direct the flow, the more gas escapes into the atmosphere and the less gas reacts at the catalyst. The thermal response of the angle cases of 30°, 45°, 60°, and 90° yielded a temperature increase of
approximately 10°C, 4°C, 1°C, and negligible temperature increase respectively. The rapid
decrease in temperature response vs. angle suggests that the gas loss to the environment became
extremely significant as the angle increased.

3.3.4. **High flow-rate experiment**

A MKS model 1480A01324CS1BM 20 SLPM maximum mass flow controller was
acquired, and it was important to characterize the behavior of the GDE at higher flow-rates in
order to implement the new mass flow controller into the transient experiments. This is important
because the transient experiments needed a higher flow-rate to compensate for the movement of
GDE. For a moving GDE, less of the gas reacts to completion; and therefore, less excitation
occurs. By increasing the flow-rate, the rate of reaction increases and compensates for the
otherwise loss heat generation. Maximizing the thermal response is important because defects
become more pronounced with high thermal response. The trade-off, however, is that conduction
to areas of lower temperature become faster with higher maximum temperatures. Flow-rates of 5
SLPM, 10 SLPM, 15 SLPM, and 20 SLPM were tested. Fluxes were also included in the legend
for thoroughness. The following experiment parameters were held constant: substrate speed of
10 fpm, orthogonal gas flow, 1 mm height between the gas knife and substrate, 2% hydrogen
concentration, and a catalyst orientation facing up. Temperature collection points were drawn
parallel to the gas knife similarly to the other parameter variation experiments. The temperature
values were averaged for each point in time. Average temperature vs. time was plotted in Figure
3.8.

![Figure 3.7: Thermal response vs. Angle](image-url)
Based on the results from the graph, it is clear that the thermal response intensifies, as the flow-rate is increased up to a certain flow-rate. It is likely that at extremely high flow-rates such as 20 SLPM, the effects of excitation begin to plateau and increases in flow-rate have less affect on temperature increase. The maximum temperature increase during testing for the 15 SLPM and 20 SLPM flow-rate cases yielded almost an identical temperature rise of 21°C, with the 15 SLPM case performing slightly better. To compensate for a faster substrate speed used for detecting defects (30 fpm), it is important to use the 20 SLPM case to deliver more gas to the catalyst. The same dips seen in the other parameter variation experiment graphs are present at time of 1 second approximately. These dips are again the result of the tape passing through the rollers and vibrating the material, which disrupts the excitation.

Figure 3.8: Temperature vs. time plot for various high flow-rates with a 2% hydrogen concentration, gas knife to substrate height of 1 mm, orthogonal flow angle, and a 10 fpm substrate speed

3.4. **Summary**

The moving substrate experiments revealed how the GDE behaves at certain conditions. The parameter variation experiments showed the behavior of the GDE by changing certain parameters. Overall, it is necessary to minimize the height between the knife and the substrate and keep the gas flow of the gas knife orthogonal to the surface of the GDE. The gas flow-rate
should be maximized as long as the temperature increases as a result. Defect detection is the next acceptable step. Detecting a defect demands proper preparation of the GDE sample and careful experimentation in order that a thermal response is generated as uniformly as possible.
CHAPTER 4 GAS DIFFUSION ELECTRODES DEFECT DETECTION

With the nature of catalytic excitation and the behavior of the GDE generally understood, it was important to begin creation of defects on the GDE in order to test defect detection using the thermal response images. A 5 mm by 5 mm defect with 100% reduction in catalyst was selected as representative of an industry encountered defect. Previous research with the IR DC method for excitation determined a 1°C difference in temperature between the substrate and the defect was adequate for detecting a defect. A substrate speed of 30 fpm was determined to be the standard testing speed for detecting defects. The following presents the progress from a general understanding of the thermal response of the GDE to detecting defects on the surface of the catalyst layer by IR thermography and RFT catalytic excitation.

In order to remain consistent, the following conditions were kept constant where necessary: gas flow-rate of 20 SLPM, 2% H₂/1% O₂ concentration, 1 mm height between the gas knife and the substrate, orthogonal gas flow to the substrate, 0.4 mg Pt cm⁻² catalyst loading, catalyst oriented up, gas knife flowing downward, and substrate speed of 30 fpm. These conditions were used for all tests unless otherwise stated.

4.1. Gas diffusion electrode sample preparation

Defects were applied to the GDE in a diagonal pattern with relatively equal spacing and were physically demarcated with notches cut into the side of the GDE to aid in processing the thermal images. The defects had approximately 100% reduction in catalyst. The reduction was determined by visually scraping off platinum catalyst from the GDE surface until the underlying GDL was completely exposed. It is likely that trace amount of Pt sill exist with in the GDL surface of the defect. A 2 mm by 2 mm square defect was later created at the same notch as the third defect in line with the first defect. A picture of the GDE sample used for defect detection experiments was provided in Figure 4.1.

4.2. Shim-stock modifier

The GDE in moving bench top roller experiments had a noticeable vibration as the material passed in between the brass rollers. This may have been caused by bumps from the tape as it passed through the brass rollers. This vibration introduced non-uniformities in thermal response videos. A plastic sheet of shim-stock with an approximate thickness of 2 mm was used during testing to add stability to the GDE. The shim-stock was flexible, but more rigid than the
GDE. The shim-stock was placed directly underneath the GDE during testing. After the shim-stock was applied, the thermal responses were uniform and strong. A picture of the shim-stock used is provided in Figure 4.2.

Figure 4.1: Gas diffusion electrode with 5mm X 5mm defects

Figure 4.2: Shim-stock used to support and stabilize GDE materials in the bench-top roller experiments.

4.3. Defect preparation

Defects were prepared manually by removing catalyst from the GDE using a laboratory scalpel knife. Defects were created as 5 mm by 5 mm square defects aligned in a diagonal fashion. Later on, a 2 mm by 2 mm square defect was applied to the GDE along the same notch as the third 5 mm by 5mm defect. The catalyst removal process did remove GDL material to the
extent that small holes were created. These holes lead to an overall change in porosity of the
defect, which may have effect detection quality. Close up pictures of the 5 mm by 5 mm defect
and the 2 mm by 2 mm defect are provided in Figure 4.3 and Figure 4.4 respectively.

Figure 4.3: Close-up image of 5mm X 5mm defect

4.4. Experiments with shim-stock

The advantage of the shim-stock was that the shim-stock improved the stability of the
GDE during testing. A potential disadvantage of the shim-stock was that flow-through to the
other side of the GDE would be restricted. This disadvantage was dismissed because excitation
occurs at the surface of the GDE, which does not require transport to the other side of the GDE.
The defects to be tested were a 5 mm by 5 mm square defect and a 2 mm by 2 mm square defect.
A defect was determined to be detected when the temperature difference between the defect and
the rest of the excited material was at least 1°C.

4.4.1. 5 mm X 5 mm defect

The 5 mm by 5 mm defects were tested along the GDE in a diagonal pattern in order to
test the GDE uniformity parallel and perpendicular to the gas knife. The camera’s zoom settings
prevented image capture of all of the defects. Only the first four could be viably seen. The
defects were individually analyzed with a line temperature probe. Frames of the first through fourth defect are provided in Figure 4.5, Figure 4.7, Figure 4.9, and Figure 4.11.

The first defect was very noticeable from the IR video frame (Figure 4.5). It is important to notice the high temperature region to the right on the GDE. This temperature region is likely a result of a vibration or bump causing the material to suddenly jolt towards the gas knife and cause excitation. It is also important to note the non-uniformity towards the bottom of the excitation on the GDE. This non-uniformity was explored in Chapter 2 and was discovered to be a result of burrs present in the gas knife holes. When the temperature line analysis is studied, the defect can be seen as the left drop in temperature. The graph is included in Figure 4.6.

The first defect is clearly visible from the line analysis graph at pixel 80 with approximate temperature drop of 2.5°C. A drop at pixel 340 can be seen because of the non-uniformity caused by the burred holes. This makes an actual attempt at detecting a defect not reliable at all. Later, post-processing analysis is used to filter out sharp non-uniformities.
The second defect is clearly visible just like the first defect (Figure 4.7). The first defect is also still visible as the second defect passes into view. Similar to the first defect, we see the same non-uniformity streak at the bottom of the frame. The hot spot present in the first defect image moved rightward, indicating that the hot spot was the result of a bump in the material and is not being systematically generated. A graph of the temperature line analysis is provided in Figure 4.8.

![Defect Detection Frame](image)

Figure 4.5: First 5mm X 5mm defect detection frame

From the second defect temperature line-analysis, we see the same general pattern of a defect drop in temperature of 2.5°C, but also with the non-uniformity streak appearing at pixel 340. Overall, reduction in noise is necessary detect a defect.

The third defect occurs at roughly the same location as the non-uniformity streak, which makes it difficult to see (Figure 4.9). The hot spot at this point in the IR video has almost completely passed through the brass rollers. A graph of the line temperature analysis is provided in Figure 4.10.
From the graph, we see that the non-uniformity streak temperature drop practically overlaps the defect temperature drop. While it is still possible to distinguish the defect drop as a more
left drop around the pixel 300, it would be practically impossible to understand a defect from a non-uniformity in the gas knife or the GDE at the location of the knife-induced non-uniformity. The temperature drop of the defect appears to be 2.5°C.

Figure 4.8: Second defect temperature line analysis

![Second defect temperature line analysis](image)

Figure 4.9: Third 5mm X 5mm defect detection frame

![Third 5mm X 5mm defect detection frame](image)
The fourth defect is positioned below the non-uniformity streak and is therefore a visible defect (Figure 4.11). The defect is clearly visible in the IR frame towards the bottom of the frame. A temperature line analysis was conducted on the defect is provided in Figure 4.12.

From the graph, we see that drop in temperature from the defect occurs to the very right of the non-uniformity at pixel 395. The defect had a drop in temperature of approximately 2.5°C. Because of the proximity of the defect to the non-uniformity streak, using the line analysis to detect the defect is unreliable.

Figure 4.10: Third defect temperature line analysis

While defects are qualitatively and quantitatively calculable, it is pertinent to refine the analysis to eliminate noise from the data and to isolate the defect. Such refining could include subtracting a baseline, non-defective area of the excited GDE, designing a more uniform gas knife, or cleaning the current gas knife. A refined subtraction analysis is demonstrated for isolating defects from a temperature analysis line. The gas knife is also cleared of burrs and cleaned thoroughly.
4.4.2. **2 mm X 2 mm defect**

As with the 5 mm by 5 mm defect, detecting the 2 mm by 2 mm defect followed the same procedure. The experiment was conducted with the exact conditions as the 5 mm by 5 mm defect detection experiment. A sample IR frame image is provided in Figure 4.13.

![Fourth defect detection frame](image)

**Figure 4.11: Fourth defect detection frame**

![Fourth defect temperature line analysis](image)

**Figure 4.12: Fourth defect temperature line analysis**
As seen in the image, the 2 mm by 2 mm is barely visible. A discontinuity in the thermal response occurs right after the defect is seen due to a vibration due the tape holding the GDL and GDE together passed through the brass rollers. The non-uniformity streak that appeared in 5 mm by 5 mm defect testing appears in the thermal video for the 2 mm by 2 mm defect. In order to gain an understanding of how well IR thermography can detect the defect, a line temperature analysis was drawn through the defect. A graph of the line analysis is provided in Figure 4.14.

![Sample frame for 2mm X 2mm defect](image)

**Figure 4.13: Sample frame for 2mm X 2mm defect**

As seen in the graph, the defect is much more difficult to point out than with the 5 mm by 5 mm defect. The drop in temperature is much less of a drop, but meets the threshold with a temperature of 1.5°C decrease. In a real detection environment, the noise and the non-uniformities could not viably detect the defect.

4.5. **Defect detection flow-rate threshold**

The purpose of this experiment was to find the flow-rate where the threshold of 1°C was met. This process was done iteratively by starting at 10 SLPM and moving to a lower or higher
flow-rate until the threshold is met. For each case, a line temperature analysis was conducted over the first defect to see the temperature drop. The threshold was discovered to be 7 SLPM right at the threshold of 1°C. A graph of the line analysis is provided in Figure 4.15.

Figure 4.14: Line analysis for 2mm X 2mm defect

Figure 4.15: Defect detection threshold temperature experiment line analysis
As seen in the graph, the defect is difficult to detect because of the noise, but has been identified at pixel 60 with a 1°C decrease from the right peak. For consistency, the flow-rate for testing should be much higher to induce a greater thermal response.

4.6. **Refined analysis of defect detection data for better results**

The purpose of this analysis was to refine the data collected for the 5 mm by 5 mm defect detection experiment in order to reduce noise and eliminate the non-uniformity streak caused the burrs present in the gas knife holes. This analysis technique could also be applied for the 2 mm by 2 mm defect detection by reducing surrounding noise. This refinement was done by identifying the IR video frame with the defect of interest (the first defect in this particular case) and then collecting the line temperature analysis data for this frame and the frame before where the line does not cross a defect. The defect line is subtracted from the pristine line. The resulting graph of this analysis line is provided in Figure 4.16.

![Temperature Difference between defect frame and base frame](image)

**Figure 4.16**: First defect detection line temperature analysis with subtraction of pristine line temperature analysis for 5 mm by 5 mm defect
In addition, the subtraction also eliminates a great deal of noise associated with the gas knife and/or the GDE. The subtraction also increased the amount of the temperature difference of the defect and the surrounding environment to approximately a 2.75°C difference. The non-uniformity streak that was at the right side of the graph is clearly reduced and has the same degree of variation as the rest of the pristine area noise.

4.7. **Deburred gas knife experiment**

Visual inspection of the gas knife holes was used to determine of machining defects contributed to the uniformity issues observed for the impinging gas flow experiments above. Trouble spots were identified in the hot plate experiment and it was important to identify visually if the holes were clogged with debris, burrs, or dirt. Holes with noticeable burrs were identified and imaged. The burred holes were then compared to relatively clean holes. The gas knife holes were then manually deburred. The cleaned holes are shown in Figure 4.17.

![Figure 4.17: Microscope analysis of gas knife of clean holes (left) and burred holes (right).](image)

The purpose of this experiment was to demonstrate the improved quality of the thermal response over previous experiments involving the gas knife due to a deburring process. This deburring process involved using a dowel pin to penetrate through each hole of the gas knife, followed by a microscope inspection of each hole to ensure the burrs were removed, followed again by a jabbing of holes with the dowel pin to finish removing burrs. The knife was then given a five-minute sonication bath in isopropanol. The sonication was conducted to remove any remaining dust or metal particles from the knife.
A transient experiment with the 0.4 mg Pt cm\(^2\) defective GDE (Figure 4.1) was conducted at the same operating conditions as with the pre-cleaned gas knife defect detection experiments. These conditions were kept the same in order to test the improvement of the cleaned gas knife over the pre-cleaned gas knife. The experiment was conducted with the material oriented forward (as indicated with the arrows on the tape) and backwards. This change in orientation was done to test to see if non-uniformities were a result of the gas knife or the material. This was repeated twice for each orientation. The results suggested that the material non-uniformities was contributing significantly to the non-uniformities seen in the thermal response. Interestingly, the backwards-thermal response generated a more uniform response and is presented below.

An IR video was taken of the transient experiment and a sample frame was taken when the first defect is very visible. As seen from the frame (Figure 4.18), the overall thermal excitation is much cleaner than the previous excitation (Figure 4.5). The non-uniformity streak that existed in the burred knife experiments is non-existent in the deburred knife experiment. Non-uniformities still exist, which may be a result of irregular hole size, or uneven spacing between holes. A line analysis was conducted on the defect and is discussed below.

Figure 4.18: Deburred knife experiment sample IR frame for first defect
A line analysis parallel to the gas knife directly through the middle of the defect was conducted on the IR image and a temperature vs. line plot was created. A subtraction was conducted similarly to the burred gas knife experiment in order to reduce edge effects and noise. The plot is provided in Figure 4.19

![Figure 4.19: Backdrop subtracted temperature vs. distance analysis of first defect for deburred gas knife experiment](image)

As seen in the plot, the defect is very visible at around pixel 80. The defect peak shows approximately a 2.5°C drop in temperature as a result of the defect. This drop is adequate for our threshold of detecting a defect. Despite the subtraction, the noise is fairly significant with approximately a 1°C amplitude for the greatest peaks.

4.8. **Summary**

From the defect detection experiments, it can be shown that a defect can be detected well within the 1°C threshold. The 5 mm by 5 mm defects for both the burred and deburred gas knife were generally detected with a 2.5°C. The 2 mm by 2 mm defect can be detected with a drop of 1.5°C. The 1°C temperature drop threshold experiments determined that a minimum flow-rate of 7 SLPM. A subtraction analysis was used to eliminate edge effects and noise for both the burred
and deburred experiments. This analysis revealed the defects in their respective temperature vs. distance plots.
CHAPTER 5 CONCLUSIONS AND FUTURE WORK

The cost of components is the primary disadvantage of the polymer electrolyte membrane fuel cell (PEMFC). In order to alleviate the costs of the PEMFCs, mass production of components is necessary. The gas diffusion electrode is a key component of the PEMFC as a part of the MEA. Manufacturing diagnostics is an important step in the manufacturing process of any product and especially the gas diffusion electrode because any defect on the catalyst layer may lead to early stack failure. In this study, we explored infrared (IR) thermography defect detection with reaction-flow through catalytic excitation using a H₂/O₂ gas mixture. A large number of experiments were conducted to characterize the diagnostic under stationary and moving environments and to optimize the diagnostic for defect detection on a moving GDE.

An array of 5 mm by 5 mm defects and a 2 mm by 2 mm defect were applied to a sample and the 5 mm by 5 mm defects were detected with approximately a 2.5°C drop in temperature. The 2 mm by 2 mm defect was detectable to within 1.5°C. The minimum flow-rate required to detect a defect was found to be 7 SLPM corresponding to a temperature change of approximately 1°C. The 5 mm by 5 mm defect was re-analyzed by subtracting the line data from line data of the same frame with no defect to get rid of noise and non-uniformities in the data. The results greatly reduced the noise and eliminated the non-uniformity streak caused by the burred holes.

The next steps of this project are the following: (1) to potentially design a more uniform gas knife for delivery of a more consistent sheet of gas to the GDE substrate; (2) to implement the diagnostic technique on a web-line with tensioned material; (3) to conduct defect testing on different level catalyst loaded GDE material; and (4) to map the catalyst uniformity on the GDE with an XRF analysis.

A uniform gas knife is necessary to ensure that the thermal response is exciting the GDE evenly. The gas knife designed for implementation in this experiment was mostly designed for a proof of concept; therefore, a low quality gas knife was justified over a more expensive commercial gas knife. In the event the RFT technique was implemented or further research is needed, a commercial gas knife would be justified.

The implementation of the technique on a web-line application would truly test the technique in an environment more realistic than the bench-top roller setup. To prepare for web-
line tests, the location of the diagnostic on the web-line would need to be determined and tested under the optimal conditions identified by this study.

Testing different catalyst loaded materials is necessary to expand the technique to the variety of loadings used for gas diffusion electrodes. Given the ultimate goal of reducing catalyst loadings on GDEs, reducing the test loading is important to justify the viability of the IR thermography technique on lower Pt loaded GDEs.

Mapping the GDE catalyst layers with XRF is important to understand where spots with too much or too little catalyst are in order to match with the thermal response videos. This characterization is mostly important for determining how close the thermal response topography matches the XRF topography in order to determine the uniformity of the gas delivery to the GDE during testing. By knowing this information, a better decision can be made regarding choosing a gas delivery source.
REFERENCES CITED


diffusion layer of low-Pt loading electrodes for polymer electrolyte fuel cells,”


