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Cold-start of parallel and interdigitated flow-field polymer electrolyte membrane fuel cell

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ABSTRACT

Achieving cold-start of polymer electrolyte membrane fuel cells (PEMFCs) from temperatures less than or equal to 0 °C can be a challenge due to the freezing of byproduct water in the cathode catalyst layer (CCL). The ability to remove water from the CCL at a rate that prevents flooding until the cell reaches temperatures above the freezing point is critical, however, they are made difficult by the low saturation pressure of air gasses below 0 °C. Under less extreme cold-start conditions (−10 °C to 0 °C) the water uptake of air can be considered non-negligible for specific flow rates and flow-field designs. Interdigitated flow-fields induce convective transport through the gas diffusion layer (GDL) and CCL under land areas, called cross flow, and have been shown to have enhanced water removal from under lands compared with parallel flow-fields. This study compares the cold-start performance between a parallel and interdigitated flow-field PEMFCs at various current densities. Testing of cold-start in an environmental chamber at −4 °C, −6 °C and −8 °C at current densities of 200 mA cm^{-2} and 400 mA cm^{-2} using galvanostatic control was completed. Results show that an interdigitated flow-field may provide better performance under less extreme cold-start conditions than parallel flow fields.

1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) must be capable of operation in extreme environments if widespread adoption of this technology is to be realized. Byproduct water is prone to freezing under sub-zero temperature conditions, which are a common occurrence in many parts of the world. Freezing water inside a PEMFC can cause reduced performance, component damage and system failure. Understanding PEMFC cold-start is a primary goal of researchers and designers that has received increasing attention over the past decade. Identifying characteristics of cell behavior under different operating conditions and flow-field designs below 0 °C is critical to enhancing cold-start capability of PEMFC technology. Methods of decreasing cold-start time, energy required for heating and degradation effects are sought after. However, very little experimental work has been conducted specifically on the flow-field design's impact on cold-start performance.

Experimental studies on cold-start have been carried out by several groups. Degradation caused by thermal cycling of PEMFCs has been reported extensively [1–6]. Damage can be attributed to stresses from residual water volume expansion in the membrane electrode assembly (MEA) during its phase transition to ice. Degradation can be reduced through the use of water removal methods including gas purging or by freeze prevention methods such as coolant loop antifreeze injection and thermal insulating [7]. Fundamental investigations into the operational effects of cold-start were performed by Oszipok et al. [8,9]. Effects of power density, gas flow rate, and membrane humidity were correlated with total current production during cold-start of single cells and stacks. Higher flow rates and lower membrane humidity increased cell survival time by allowing for water uptake and removal from the catalyst layer (CL). Yan et al. performed a study of sub-zero temperature effects on PEMFC performance and components [10]. They showed successful start up from −5 °C if the cell was properly insulated and pre-purged. Higher flow rates and feed gas temperature made it possible to start from −10 °C using a step current density of 100 mA cm^{-2}. Scanning electron microscopy showed damage to MEA layers, including delaminating of the CL. Chacko et al. used high-frequency resistance measurements to characterize a membrane undergoing cold-start from −10 °C [11]. They found high current densities produced a higher heating rate but a lower total amount of heat before failure. Tajiri and Wang performed isothermal cold-start testing showing that higher current densities lead to incomplete utilization of cathode catalyst layer (CCL) pore space [12].

Fundamental studies on cold-start focused on understanding the water transport process under freezing conditions. Cell failure during cold-start can be attributed to the flooding of the CCL with

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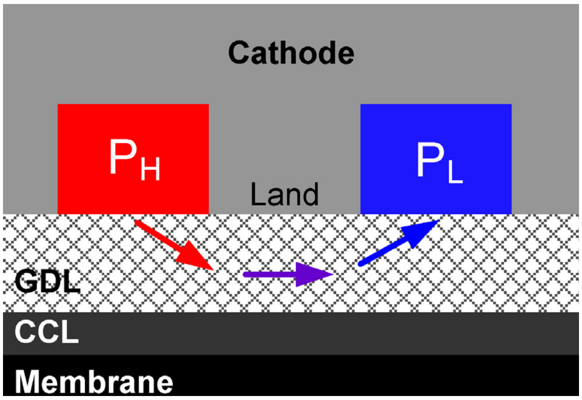

Fig. 1. Diagram of cross flow between adjacent channels. P_H represents a high pressure channel and P_L a low pressure channel. Arrows designate transport beneath the land area through the GDL.

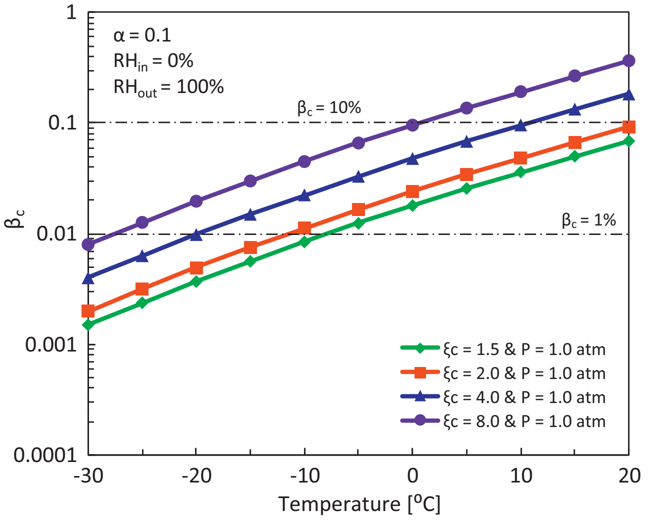

Fig. 2. Chart based on the methods of Wang, using the conditions for this cell testing, β_C is plotted on a log scale. The temperature range of interest for this study is between −10 °C and 0 °C, which are less extreme examples of cold-start resulting in β_C greater than 1%.

water as low temperatures make water removal difficult. If flooding of reaction sites outpaces heat production, water may freeze causing a drop in cell performance and cell failure. Ge and Wang imaged liquid water formation under sub-zero operation and identified a freezing point depression of 1 ± 0.5 °C in the CCL [13,14]. Mao and Wang developed a 1D analytical model for cold-start which was later integrated into a 3D multiphase model [15,16]. They identified the ability to remove water from the CCL at a rate that prevents flooding until the cell reaches temperatures above freezing as paramount. Their multiphase model predicted a non-uniform water distribution between channel and land areas during cold-start with a propensity for higher ice fractions occurring under land regions. Wang expanded on the analytical analysis of CCL transport phenomena assuming convective water transport, inherent in interdigitated flow fields, as a primary water removal mechanism from the GDL [17]. At sub-zero temperatures it was shown that cold air has a lower capacity to hold water and that convective water transport may be considered negligible at lower flow rates.

Several computational modeling and simulation studies have examined cold-start phenomena [18,19]. Meng concluded that increased gas flow rates in the cathode region are beneficial to cold-start performance [20]. His study found lower current densities produced less water which prolonged cell lifetimes. However, he noted practical cold-start times would require higher amounts of waste heat production usually achieved at higher current densities. Results also showed higher levels of water present under land areas. Jiao and Li completed extensive numerical based cold-start analysis under varying conditions [21–23]. One key finding was that although low current densities upon start up allow for higher pore space utilization, higher current densities produce more waste heat per water created and therefore may be beneficial to cold-start. Due to the land wall heat conduction, ice was shown to appear first under land areas. This study showed minimal benefits to heating incoming gasses and that high gas flow rates would be needed to improve water removal. Recently, Ko and Ju used a multiphase model to investigate ice formation in the CCL based on varying initial water contents [24]. They confirmed lower initial water content increases cell survival time. Results showed that higher ice fractions originating under land areas may be a result of higher water production due to higher current densities initially under land areas in addition to lower temperatures in these regions.

The primary difference between parallel and interdigitated flow fields is that under parallel flow conditions, water removal from the MEA is dependent upon diffusion, while interdigitated flow conditions induce convective transport [25,26]. Extensive non cold-start water management studies were completed by Trabold and Owejan et al. using neutron radiography (NR) examining the effects of serpentine and interdigitated designs [27,28]. Spernjak et al. used NR to compare water transport characteristics between parallel, single-serpentine and interdigitated flow-field [29]. Results showed that interdigitated flow fields were more effective in removing product water from cathode land areas compared to parallel and serpentine patterns. Convective flow over land areas is referred to as cross flow, displayed in Fig. 1, and has been studied by Li's group [30–32]. Their findings demonstrate the ability of cross flow to reduce concentration over potential by enhancing delivery of reactants and removal of product water under land areas. Such water removal capabilities may be beneficial to PEMFC cold-start performance since land areas are susceptible to lower temperatures during cold-start and are thus sensitive to water build-up/freezing in the CCL. Recently, in-plane NR of cold-start carried out by the Paul Scherrer Institute confirmed the presence of water/ice buildup under land areas [33]. Papadias et al. mentioned possible benefits of cross flow to cold-start, performing preliminary simulations of interdigitated sub-zero water transport which showed the potential for improved performance [34]. This paper will compare parallel flow-field performance to interdigitated flow-field performance during cold-start in order to investigate the effects of cross flow.

2. Testing parameters and data analysis

In order to compare the performance of parallel and interdigitated flow field cold-start, the effect of the design on key variables will be analyzed. It is recognized that at sub-zero temperatures, water removal is difficult because of its low saturation point which means use of high gas flow rates may be needed [21]. Conditions of interest for this study are those where the ratio of gas stream water removal to CCL water production is not negligible which requires higher flow rates and temperatures. Wang derived a dimensionless parameter which represents the ratio of water gained by the cathode gas flow stream to the amount water produced in the CCL accounting for membrane water uptake and back diffusion for an interdigitated flow-field PEMFC [17]. This parameter, β_c, is defined as:

$$\beta_c = \frac{\xi_c(RH_{c,\mathrm{out}}C^{\mathrm{sat}}_{\mathrm{out}} - RH_{c,\mathrm{in}}C^{\mathrm{sat}}_{\mathrm{in}})}{2(1+2\alpha)\,C^{O_2}_{c,\mathrm{in}}} \qquad (1)$$