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Investigation of time dependent water droplet dynamics on porous fuel cell material via synchrotron based X-ray imaging technique

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ABSTRACT

Proper water management in proton exchange membrane fuel cells (PEMFCs) remains a challenge, hindering the commercialization of this technology. Liquid water emerges in the form of droplets from micrometer-scaled pathways of porous gas diffusion layers (GDLs) into millimetre-scaled gas flow channels. Afterward, droplets grow to certain critical sizes before being removed by the convective gas flow. The opaque nature of fuel cell components, including GDL materials, poses significant challenges to understand droplet dynamics and develop more efficient water management techniques. In this work, an advanced synchrotron X-ray radiographic imaging technique available at the Canadian Light Source (CLS) was employed to visualize time dependent droplet behaviour in a serpentine PEMFC flow channel for the first time. High spatial and temporal resolution coupled with high energy photons of an X-ray beam provided high resolution images of growing droplets. From the captured images, the whole growth cycle of liquid droplets was obtained under various gas flow rates. A cyclic pattern of droplet dynamics including emergence, growth, and detachment was visualized from high resolution images. From the X-ray images, the time-evolution of dynamic contact angles, height, and chord of a growing droplet was quantitatively analyzed. At higher gas velocities, the critical detachment diameter of droplets became smaller and contact angle hysteresis was found be lower, while larger contact angle hysteresis and bigger droplets were observed at lower gas velocities. The critical droplet size at which the detachment of droplets occurred was compared to the predicted results from an analytical approach and good agreement was found.

1. Introduction

Water is produced initially at the cathode catalyst layer of proton exchange membrane fuel cells (PEMFCs). After being pushed through the interconnected water pathways of porous gas diffusion layers (GDLs), liquid water can develop into droplet, film or slug flow regimes in the gas flow channels of the fuel cell depending on the flow conditions [1]. Since excess water can result in flooding and negative impacts on the fuel cell's performance, high gas flow, typically air, in the cathode flow channels is needed to purge liquid water out of the cell. However, high superficial gas velocities result in significant parasitic power losses [2]. In order to find the optimal condition for removing excess water from the gas flow channels, a systematic study revealing how changing superficial gas velocity can influence liquid water behaviour is of great importance. Despite the importance, due to optical inaccessibility of both the GDL and gas flow channels, droplet evolution from emergence to detachment is still not well understood.

In terms of theoretical attempts and modeling, several research groups conducted studies on parameters controlling the formation and detachment of droplets [1–5]. Theodorakakos et al. [6] investigated the droplet surface deformation to predict the actual shape of droplets sitting on three different GDLs using static and dynamic contact angles of droplets as an input for the volume of fluid (VOF) method. They indicated that droplet geometry altered dramatically from the initial static shape up to the point of detachment. Chen et al. [7] assumed spherical droplets to present their simplified model based on macroscopic force balances and compared the results with experimental visualization data at low and high gas velocities. However, measuring the droplet aspect ratio at different gas velocities, Wu et al. [8] concluded that assuming a spherical shape for growing droplets in simplified models is very approximate due to hydrodynamic forces exerted by the gas flow. Zhu et al. [9] simulated the behaviour of droplets emerging through a micro-sized pore, which was followed by droplet growth and its interaction with a hydrophobic surface. Utilizing the VOF method,

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they investigated the effect of water connectivity inside a micro-sized pore on critical detachment air velocity, which was significantly lower for the stagnant droplets sitting on the surface without pore pinning. However, information obtained from simulated gas flow channels as reported in literature is different than actual conditions for operating fuel cells in several ways. In general, simulations in this field lack rigorous experimental validation. For instance, the pore is often imagined as a single injection port and the simulated surface does not exactly represent a true GDL [10–13]. Further, while a contact angle of a wall can be set, the models often do not include true variation in the contact line that can occur over time at the interface [13].

In terms of experimental attempts, the opaque nature of various cell components poses significant challenges for visualization of droplet behaviour at the cathode side of PEMFCs. Bazylak [14] presented a review paper on strengths and drawbacks of different visualization methods utilized in fuel cell studies including neutron imaging [15–20], NMR imaging [21–25], optical photography [11,12,26–31] and synchrotron X-ray imaging [32–34]. Minor et al. [35] utilized micro-digital-particle-image-velocimetry (micro-DPIV) in optical photography to correlate air velocity with droplet deformation and contact angle hysteresis. They concluded that side-view optical images from this method were not adequate to obtain concrete information regarding true interaction between the air-water interface for the air phase. Thus, simultaneous top-view images were required as well. Using an optical visualization technique, Mortazavi et al. [36] found that due to the interconnected network of water pathways within GDLs, droplet emergence locations were totally random. In contrast, Yang et al. [37] pointed out that there were certain preferential locations for emerging droplets in gas flow channels. Bazylak et al. [38] concluded that preferential pathways existed, but these could shift over time, making it appear dynamic or random over longer timescales.

Although optical visualization of droplet behaviour could be possible through a clear plate from the top-view, capturing the evolution of a growing droplet in terms of dynamic contact angles and critical droplet height is required to be from the side. Considering the preferential emergence of droplets in random locations of serpentine or parallel channels, side-view optical visualization is not feasible due to channel overlapping in parallel or serpentine flow fields. Also, different flow regimes that occur simultaneously along the serpentine channels due to non-uniform water emergence between the inlet and outlet of long channels cannot be captured separately for the same reason [39].

Synchrotron based X-ray imaging as a result of high energy penetrating photons plus high spatial and temporal resolution makes the observation of multiphase flow within the length of a serpentine channel feasible. Manke et al. [41] used synchrotron X-ray imaging techniques to investigate liquid water evolution and transport through GDLs in gas flow channels of an operating fuel cell. Based on a quantitative analysis, they found a dynamic equilibrium between droplets in the channel and water clusters in the GDLs. Fluckiger et al. [42] studied liquid water saturation in the porous GDL network using X-ray tomographic microscopy and found that liquid water saturation depends on water pressure, indicating the retention effect of liquid water at gas diffusion layers with higher PTFE loading level. Roth et al. [43] characterized the structure of a dry GDL and investigated the in-situ characteristic length scale of liquid water across the GDL employing the concept of representative equivalent area (REA). Utilizing X-ray imaging, they noted that the local water distribution pattern is mainly dominated by the micro-scale structure of the GDL since the water distribution pattern re-formed after drying and humidifying again. Pasaogullari et al. [44] proposed one-dimensional analytical solutions to predict the onset of two-phase flow in both hydrophobic and hydrophilic GDLs depending on different relative humidity combinations. Analyzing the influence of GDL hydrophobicity on liquid water transport, they revealed that liquid water transport across the GDL is mainly dominated by capillary forces. Eller et al. [45] investigated water saturation variation of the GDL and cell performance deterioration after synchrotron irradiation. Lee et al. [40] employed synchrotron X-ray radiography to compare the liquid water transport through GDLs with and without microporous layers (MPL). They reported that GDLs assembled with an MPL could prevent flooding due to the nano-scaled pores of the MPL, which could result in smaller water clusters and lower water saturation in the GDL.

It can be seen that though attempts of using X-ray radiographic imaging techniques have been made to understand liquid transport in PEMFCs, specific attention on time dependent droplet behaviour, particularly droplet growth cycles and dynamic contact angles, remains very limited. The dynamic contact angle of droplets during the droplet flow regime is a key factor for fuel cell water management, and it is determined by interfacial energy along the three-phase boundary. Drag force, surface adhesion force, and capillary pressure have an influence on the droplet's dynamic contact angles on the GDL surface. The surface adhesion force resulting from solid-liquid molecular interactions makes growing droplets adhere to a solid surface by overcoming the drag force imposed by the gas flow [36]. Nevertheless, drag force makes the advancing and receding contact angles of the droplet different from the static contact angle, and the resulting difference between advancing and receding contact angle is termed "contact angle hysteresis". Kumbur et al. [5] noted that contact angle hysteresis was a function of droplet size, superficial gas velocity, and contact surface properties. In particular, dynamic contact angles and contact angle hysteresis are much needed information for numerical modeling and simulations where static contact angles are often used. Therefore, in view of this knowledge gap, the main objectives of the current work are to understand time-dependent droplet dynamics through advanced X-ray radiographic imaging techniques and to provide information on dynamic contact angles during droplet growth. The focus of this study is on how droplet hysteresis is (1) varying with time and (2) cyclical. This work shows how long it takes for a droplet to reach certain advancing and receding contact angles, and how this process is noted by multiple, distinct 'phases' of contact angle behaviour before detachment. To the point above, this behaviour is not analyzed in CFD simulations. In addition, the experimental results of critical droplet sizes are compared to those predicted from an analytical approach previously developed [8]. The present work provides valuable insight into the evolution and instability of growing droplets and suitable quantitative data to verify numerical analyses and theoretical models.

2. Experimental section

The ex-situ fuel cell used in this work was designed for synchrotron X-ray visualization of experiments on dynamics of liquid water at the cathode side of a fuel cell. In-plane X-ray visualization was conducted while the direction of the X-ray beam was horizontally parallel to the GDL plane. The following sections detail the test cell design, experimental conditions, and synchrotron radiography setup.

2.1. Microchannel design

The microfluidic device (i.e. ex-situ fuel cell) fabricated specifically for these experiments includes a serpentine channel with dimensions of 0.5 mm × 1 mm (height × width). The reason for opting for a serpentine channel was to increase the probability of capturing droplet growth cycles in different parts of the gas channel. To represent the actual droplet emergence phenomena relevant to PEMFC operation, instead of using a single water injection pore, liquid water emerges from a Sigaret 25 BC GDL ensuring the pore size and emergence locations are randomly distributed. The uncompressed, 235 μm thick GDL is loaded with 5%wt polytetrafluoroethylene (PTFE), assembled with a MPL, and has a reported air permeability of 1 $cm^3/(cm^2\,s)$.

Fig. 1 shows the acrylic test cell with a single serpentine minichannel with a length of 35 mm simulating an operating PEM fuel cell with active area of 132 mm^2. Acrylic was selected as it minimizes X-ray