



Gas management and multiphase flow in direct alcohol fuel cells



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ABSTRACT

Direct Alcohol Fuel Cells (DAFCs) have the potential to compete with current battery technology due to the high energy density of the alcohol fuels. A major limitation to these fuel systems is “blinding” where CO₂ gas pushes through the porous gas diffusion layer (GDL) forming bubbles in the flow channels inhibiting fuel delivery to the catalyst layer. We report here measurements of material wettability, advancing and receding contact angles, and gas bubble formation to describe multiphase transport processes relevant to DAFCs. Carbon paper and carbon cloth have large differences in the receding contact angles ($\theta_{R,paper} = 85^\circ$, $\theta_{R,cloth} = 120^\circ$), which result in differences in bubble growth and detachment from the GDL surface. Bubbles on carbon paper were ~40–50% smaller than on carbon cloth. The gas pressure to push gas through the GDL was 50–80% greater for carbon paper. The geometry of the flow channel also played a major role in confining bubble growth. The permeable nature of the GDL resulted in lateral transport of gas to existing bubbles. This fundamental understanding of transport processes suggested an electrode design with dedicated gas removal channels that could enable a DAFC to operate without bubble formation.

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1. Introduction

Direct liquid (ethanol, methanol, formic acid) fuel cells generate carbon dioxide as a product of the anode reaction. Gaseous product builds up in the catalyst layer and pushes through a porous electrode (the gas diffusion layer) into the liquid flow channel forming bubbles [1–4]. These bubbles are moved along the flow channel and out of the fuel cell. Gas pockets in the fuel cell, either within the GDL/catalyst layer or in the flow channel, block liquid fuel delivery to regions of the fuel cell and decrease effective power density [5–8]. Gas management has been extensively studied and efforts have been explored to minimize the impact on performance [1–4,7–9].

Engineered solutions to enhance gas removal include flow channel geometry [3,7,8] and gas diffusion media selection [9], dissolution of the gaseous product via increased pressure [10] or chemical agents [11], and in situ membrane separations [4]. Most efforts have heuristically correlated cell voltage or current to changes in materials or operating conditions (temperature, flow rate). We are focusing on developing a fundamental understanding of the surface wetting properties of the materials relevant to direct alcohol fuel cells and the impact those properties have on multiphase transport processes. Previous engineering approaches

to gas management examined various flow channel designs and characterized their performance in situ, justifying better performance with various assumptions [2,5,12]. To this point in time there has been very little connection between fundamental design of the channels (wettability, geometry) and transport processes. Our objective is to improve our physical understanding of gas/liquid flow in porous media and microchannels to provide the scientific basis for designing electrodes for direct alcohol fuel cell systems.

These multiphase flow issues at the anode are the inverse of the liquid water management dilemma at fuel cell cathodes. In hydrogen fuel cells, liquid slugs form blocking gas transport from the gas flow channel to the catalyst/membrane interface. In liquid fuel cells carbon dioxide bubbles form and block liquid fuel transport. To isolate the fundamental physics of multiphase flow relevant to hydrogen fuel cells we have employed simplified microfluidic systems [13–16]. The ultimate goal is to demonstrate how the structure of the porous electrodes and geometrical dimensions of the flow field control the growth and motion of bubbles within fuel cells. By understanding the fundamental physics of the multiphase flow and the influence of material properties it will be possible to design and control the fuel cell for enhanced performance. It is our belief that design strategies could be utilized to achieve improved gas management resulting in increased power density. In this paper we will discuss our work on multiphase relevant to direct alcohol fuel cells and the carbon dioxide gas management issue.

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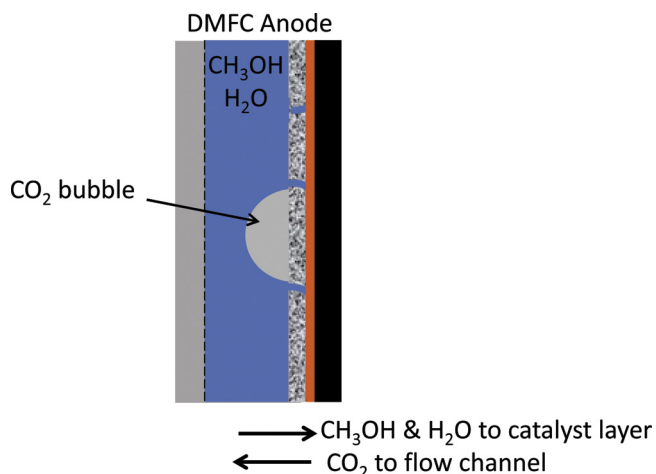
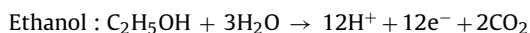
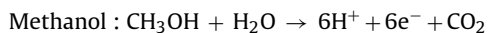


Fig. 1. Schematic of multiphase transport in direct methanol fuel cell anode.

1.1. The carbon dioxide management issue

Direct alcohol fuel cells generate carbon dioxide at the anode via the following anode reactions:



There are additional partial oxidation products that will result in differences in gas production. Electrocatalysis research efforts try to promote complete oxidation. In our analysis we will assume complete oxidation to carbon dioxide with the acknowledgment that this is not always true. As shown schematically in Fig. 1, a mixture of liquid water and alcohol must diffuse to the anode catalyst layer while CO₂ is transported away. Initially, the CO₂ saturates the dilute fuel solutions (methanol is typically a 1 M solution [17–20]). Solubility of CO₂ in water is low so the CO₂ produced will saturate the solution and form gas bubbles.

Typically, direct alcohol fuel cells use a conventional hydrophobic GDL which leaves a majority of the material open for gas-phase transport while relying on a combination of liquid delivery through large pores and diffusion of methanol through the gas-phase regions [3,9]. This situation results in diffusional limitations that may be alleviated by a more hydrophilic GDL. Zhang et al. [9] treated a GDL with a microporous layer of variable wettability and found that the more hydrophilic treatment resulted in the highest power density.

Several efforts have examined how flow channel design and fuel cell operation can facilitate bubble formation and removal to improve fuel cell performance. As expected, increased liquid flow rate facilitates the removal of gas bubbles at the expense of higher parasitic losses through increased pump capacity and decreased alcohol conversion [7]. Yang and Zhao used a transparent fuel cell anode to demonstrate that a single serpentine flow channel design results in less gas accumulation and higher power output than a parallel flow channel system. In the serpentine channel, the higher flow rate effectively cleared the gas blockages but resulted in a higher pressure drop. Flow will divert through different channels to avoid a blockage in the parallel flow field design.

Meanwhile, Oliveira et al. [3] concluded that the single serpentine channels resulted in too large of a concentration gradient along the length of the flow channel. Reactants are depleted and by the end of the channel the stream is very dilute resulting in low power output. They suggested a mixed flow channel design with small pressure gradients. They did not have visualization capability to compare the gas accumulation in the different configurations.

While the concentration gradients are relatively straightforward to characterize in a fuel cell, multiphase transport presents a more complicated problem that has not been sufficiently explored from a fundamental basis.

2. Experimental

We have devised experiments intended to mimic the transport processes relevant to direct alcohol fuel cells. These experiments provide more insight about multiphase transport processes than can be observed in an operating fuel cell. The results are useful for informing electrode design and operation strategies.

2.1. Materials

The gas diffusion materials tested were purchased from Fuel Cell Earth LLC. Toray carbon paper materials (TGP-H-120) were obtained with treatments of 20 and 40 wt% Teflon and a thickness of 370 μm. Woven carbon cloth was supplied by Fuel Cell Earth with 20 and 40 wt% Teflon and a thickness of 380 μm.

The GDL is a material with complicated pore and surface structures. Porous media are frequently modeled as arrays of cylindrical pores. We sought to understand the difference between transport in the GDL and transport through cylindrical pores by fabricating an array of pores in a polyimide sheet. The polyimide standard was fabricated with 5 laser drilled pores of 100 μm diameter as shown in Fig. 2. The 25 μm thick polyimide was purchased from McMaster-Carr.

2.2. Bubble formation experiments

A GDL was positioned in a polycarbonate block with silicon rubber gaskets was used to hold and seal the material under investigation. Liquid water was above the GDL and a flowing gas was supplied from below the GDL. Gas diffusion materials are impermeable to liquid water for hydrostatic pressures of several centimeters of liquid water as reported previously [13,15,21]. Typically the water height above the sample was 2 cm. The volumetric flow rate of N₂ gas into the lower chamber was controlled using a rotameter (Matheson FM-1050S-V). The pressure was monitored in the lower gas chamber using a pressure transducer (Omega PX164-005D5V).

The orifice pressure reported in this work is the difference between the measured pressure in the gas chamber and the hydrostatic pressure.

$$\Delta P_{\text{orifice}} = \Delta P_{\text{measured}} - \Delta P_{\text{hydrostatic}} \quad (1)$$

2.3. Channel flow

The channel flow experimental setup was the same as that developed by Colosqui et al. [14]. 1.6 mm² by 125 mm long channels were machined in an acrylic block. A 0.1 mm hole was drilled through the acrylic centered in the channel at a distance of 102 mm from the end of the channel. We performed experiments on this setup both with and without GDL on the surface in contact with the 0.1 mm pore. A 20 wt% Teflon treated Toray carbon paper (TGP-H-120) GDL was held in position with a very thin layer of silicon grease to isolate gas flow through the GDL (not allowing flow at the interface between the acrylic block and the GDL). We recorded video images and time correlated those with pressure traces of both the liquid and gas feed using Omega PX-160 series transducers. The primary distinction in these experiments versus those by Colosqui is that the gas and liquid are reversed. Liquid is the primary feed into the flow channel while gas is fed through the 0.1 mm diameter side channel hole. Gas flow is controlled by an AALBORG mass

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