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Analysis of Pt/C electrode performance in a flowing-electrolyte alkaline fuel cell

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ABSTRACT

We characterize the performance of Pt/C-based electrodes under alkaline conditions using a microfluidic H₂/O₂ fuel cell as an analytical platform. Both anodes and cathodes were investigated as a function of electrode preparation procedures (i.e., hot pressing, acclimatization) and fuel cell operating parameters (i.e., electrolyte composition) via chronoamperometric and electrochemical impedance analyses. X-ray micro-computed tomography was employed to link electrode structure to performance. In addition, the flowing electrolyte stream is used to study the effects of carbonates on individual electrode and overall fuel cell performance. Our studies provide direct evidence that the performance of hydrogen-fueled room-temperature alkaline fuel cells (AFCs) is limited by transport processes to and from the anode primarily due to water formation. Furthermore, the presence of carbonate species in the electrolyte appears to impact only anode performance whereas cathode performance remains unchanged.

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1. Introduction

For the widespread commercialization of present acidic polymer electrolyte membrane (PEM)-based fuel cell technologies to be realized, significant enhancements in durability and reductions in cost are needed [1,2]. Of late, alkaline fuel cells (AFCs) have received renewed interest because they may overcome some of the hurdles that hamper the commercialization of their acidic counterparts. Under alkaline conditions, the kinetics of the oxygen reduction reaction (ORR) on the cathode are enhanced leading to improved fuel cell energetic efficiency and reduced need for high loadings of precious metal catalysts, i.e., platinum (Pt) [3]. Furthermore, a wider

range of materials are stable in alkaline media, compared to acidic media, facilitating the implementation of cheap non-noble metal catalysts, i.e., silver (Ag) cathodes and nickel (Ni) anodes, as well as other low-cost structural materials, i.e., Ni current collectors, which can significantly lower fuel cell costs [4]. Consequently, AFCs have significant promise as an alternative to acidic PEM-based fuel cells for low-temperature applications [5,6].

Traditional AFC configurations with stationary liquid electrolytes, typically concentrated potassium hydroxide (30–45 wt% KOH), have been applied successfully over many decades, for example in vehicular applications and in the NASA Apollo space program in the 1950s and 1960s [1,5,7,8].

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The major technical concerns for these traditional AFCs are (i) electrode durability in highly caustic environments (i.e., hydrophobicity losses), (ii) carbonate formation when oxidizing organic fuels directly, and to a lesser extent (iii) water management at the electrodes (anode flooding and cathode dry-out) [9]. Of particular importance is carbonate formation ($\text{CO}_3^{2-}/\text{HCO}_3^-$), which occurs when the hydroxyl ions (OH^-) present in the electrolyte react with carbon dioxide (CO_2) mainly from either organic fuel oxidation, or the environment in which the system operates (e.g., air, tailpipe emissions from automobiles). In the presence of mobile cations, the carbonates can precipitate within the electrodes, where they damage and/or clog the microporous architecture, block electrocatalytic sites, and eventually reduce performance. Furthermore, carbonate formation reduces the OH^- concentration in the liquid electrolyte, thus lowering electrolyte conductivity and electrode reaction kinetics. Consequently, these traditional AFCs were mainly limited to applications where cost was not a concern, which allowed for the use of high purity hydrogen (H_2) and oxygen (O_2) gas streams with ultra-low CO_2 concentrations and no internal carbonate formation by definition, thus avoiding carbonate formation-related issues in the stationary electrolyte [7].

To alleviate carbonate and water management-related issues, AFCs with circulating electrolyte streams have been developed that exhibit improved performance and lifetime compared to AFCs with stationary electrolytes [4,9–12]. The flowing electrolyte improves heat and water management and facilitates carbonate removal, which results in CO_2 tolerance up to ~ 100 ppm [9]. While such flowing configurations prolong AFC lifetimes, the electrolyte solution must still be periodically replenished or replaced to maintain conductivity and prevent carbonate precipitation due to saturation over long operational lifetimes. To further extend lifetimes, modern liquid electrolyte-based AFCs also incorporate CO_2 scrubbers (i.e., soda lime) at the air inlet. However, the circulating electrolyte system and scrubber are ancillary systems that increase both parasitic losses and device complexity. In addition, electrode durability in the caustic environment remains a challenge [9]. Leaking of concentrated electrolyte has also been cited as a consumer safety concern [4].

As an alternative to liquid electrolyte-based AFCs, novel alkaline anion exchange membrane (AAEM)-based fuel cells are being pursued because these membrane-based designs reduce system complexity and increase device robustness but still maintain the electrocatalytic advantages of operating under alkaline conditions [13–16]. Furthermore, AAEMs are less susceptible to carbonate precipitation because no mobile cations exist within the membrane enabling less stringent operating conditions such as direct air-breathing cathodes. Still, the presence of carbonate ions in or near the AAEM can adversely impact cell performance (e.g., unfavorable pH gradients, reduced conductivity) particularly in the case of direct liquid AAEM-based fuel cells [17]. Also, carbonate precipitation is still possible in the presence of metal ions in the electrode structure or the reactant streams [2]. Over the past decade, research on AAEM technologies has led to dramatic enhancements in cell stability and membrane conductivity [18]; however, several key challenges remain such as high costs and insufficient performance and durability

under fuel cell operating conditions, especially at elevated temperatures [13]. For example, AAEM-based fuel cells, like acidic PEM-based fuel cells, are hampered by membrane limitations, notably membrane conductivity and water management at the electrodes (anode flooding and cathode dry-out) [19].

Developing a better understanding of the complex electrochemical, transport, and degradation processes that govern the performance and durability of electrodes within operating fuel cells is critical to designing the robust, inexpensive configurations that are required for commercial introduction [20–23]. However, detailed *in-situ* investigations of individual electrode processes are complicated by other factors such as water management, uneven performance across electrodes, and temperature gradients. Indeed, too many processes are interdependent on the same few variable parameters, necessitating analytical platforms with high degrees of freedom. To this end, we have developed a pH-flexible microfluidic H_2/O_2 fuel cell with a flowing electrolyte stream instead of a stationary membrane (Fig. 1) [24–26]. The flowing electrolyte (i) minimizes adverse fuel cell system limitations, i.e. water management, (ii) enables independent control of electrolyte parameters (i.e., pH, composition) and consequently local electrode environments, and (iii) allows for *in-situ* studies of single electrode performance via an external reference electrode [25,27]. Previously, we have demonstrated the utility of this analytical platform to characterize the performance of Pt/C and Ag/C-based cathodes under alkaline conditions [24]. This platform can be used to systematically probe the key structural factors and operating conditions that govern the performance and durability of individual electrodes within operating AFCs. Such studies will be critical in guiding the development of robust and cost-effective liquid- and membrane-based AFC systems.

Here, we use the microfluidic H_2/O_2 fuel cell to identify performance-limiting factors for room-temperature H_2 -fueled AFCs. The performance of Pt/C-based electrodes, both anodes and cathodes, is investigated as a function of electrode preparation protocols (i.e., hot-pressing, acclimatization) and cell operating parameters (i.e., electrolyte composition). In addition, we analyze the impact of carbonate species on individual electrode and overall fuel cell performance by introducing the contaminants via the flowing electrolyte stream.

2. Experimental

2.1. Gas diffusion electrode preparation

For each electrode, a catalyst ink was prepared by mixing 8 mg Pt/C (50% mass on Vulcan carbon, E-Tek), 5.33 mg polytetrafluorethylene powder (PTFE, Aldrich) as the hydrophobic catalyst binder, 200 μL Millipore water (18.2 M Ω) and 200 μL isopropyl alcohol. Previously, the optimal weight percentage of PTFE to the total weight of the PTFE/catalyst mixture within the catalyst ink was determined to be 40 wt% [24]. This catalyst ink was sonicated (Branson 3510) for 1 h to obtain a uniform mixture, which was then painted onto the microporous side of a Toray carbon paper gas diffusion layer (GDL, EFCG “S” type electrode, E-Tek). The GDLs consist of two

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