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Carbon Corrosion in Polymer Electrolyte Fuel Cells: A Complex Interplay between Morphological Changes and Electrochemical Performance

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Abstract

Due to the high gravimetric energy density of hydrogen, the focus of implementation of polymer electrolyte fuel cells (PEFCs) has shifted from light duty passenger vehicles to heavy duty vehicles such as buses, trucks, locomotives and marine vessels[1]. A mechanistic understanding of degradation is therefore necessary to improve durability and efficiency. During start-up and shut-down (SUSD) of PEFC systems, the catalyst (Pt nanoparticles embedded on carbon support) undergoes local potentials ~ 1 - 1.5 V caused by a combination of fuel (H_2) starvation, mixed fuel region and cell reversal[2]. This leads to a series of degradation phenomenon including reduction in cathode catalyst layer (cCL) thickness and porosity, loss in electrochemical surface area (ECSA), ionomer degradation and loss in electrical contact, therefore resulting in severe performance loss[2]. The convoluted relationship between these individual degradation mechanisms, their chronology and their effects on electrochemical performance are yet unresolved.

In this study, the complex interplay between morphological changes due to carbon corrosion and its effects on the electrochemical performance were analyzed using a combination of detailed electrochemical characterization, spectroscopy, and electron microscopy techniques. Commercially available catalyst coated membranes were subjected to the Department of Energy carbon corrosion accelerated stress test (AST) protocol. It was found that carbon corrosion in PEFCs can be divided into two phases. In the first phase (~500 cycles), disordered domains of the carbon containing Pt nanoparticles oxidized faster with a severe loss of cCL thickness. Pt nanoparticles detached from the support and became electrically isolated and hence electrochemically inactive. Electrochemical surface area decreased rapidly in this phase with little change in the cCL pore structure. Both kinetic and mass transport overpotentials (due to decrease in roughness factor) increased. In the second phase (remaining 1500 cycles), a severe pore structure collapse was observed (see Figure 1). Porosity, mean pore size, and the pore

connectivity decreased considerably. Pressure independent oxygen mass transport resistance showed anomalous increase. The oxygen-reduction reaction kinetics were impeded, reflected by an increase in the Tafel slope. This was caused due to severe mass transport limitation, even at low current densities of 0.1 $A.cm^{-2}$ in oxygen environment owing to pore structure collapse. In addition, the AST did not change the concentration of oxides on carbon, however, the overall water management in the cCL deteriorated as the pores lost percolation. Ionomer degradation was observed in the second phase. Sheet capacitance and sheet resistance also decreased with carbon corrosion.

Figure 1. a) 3D reconstructions of the cCL using FIB-SEM micrographs, b) porosity as a function of thickness. Scale bar is 1 μm.

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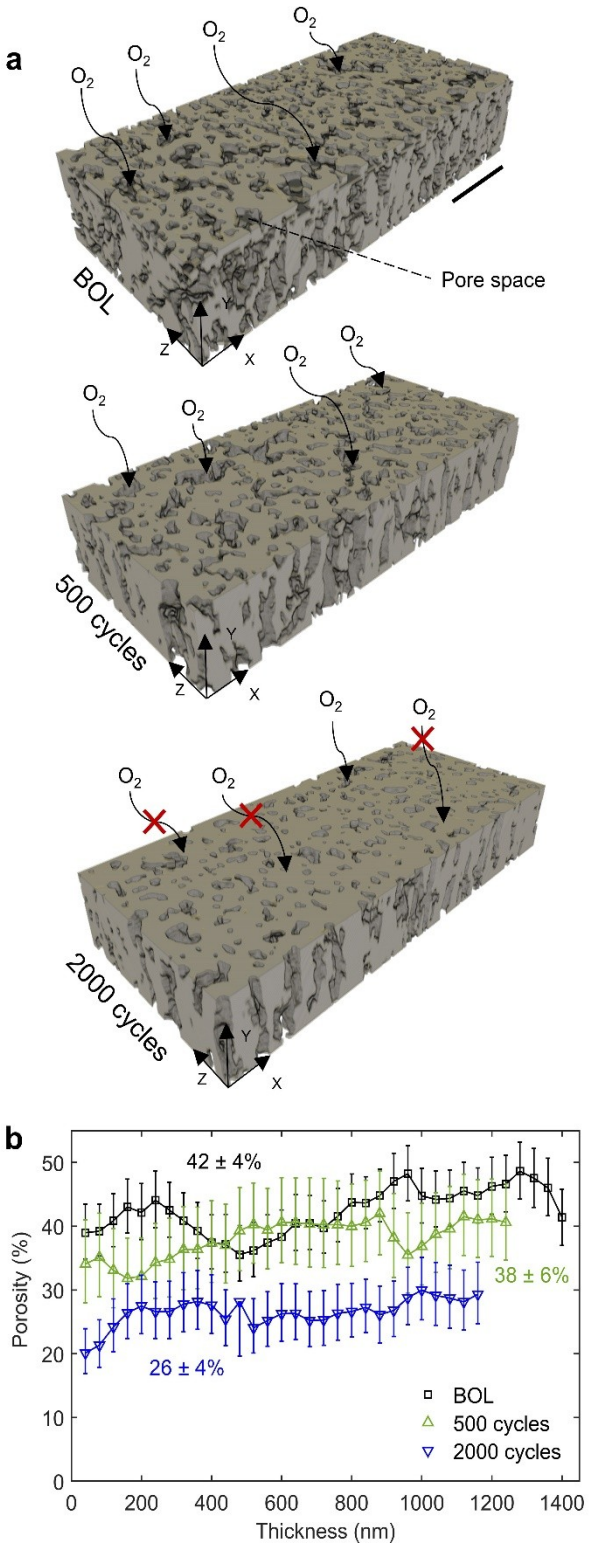
a
O2
Pore space
BOL
Y
Z
X
500 cycles
2000 cycles
b
42 ± 4%
38 ± 6%
26 ± 4%
BOL
500 cycles
2000 cycles
Porosity (%)
Thickness (nm)
0
10
20
30
40
50
0
200
400
600
800
1000
1200
1400

Figure 1