Introduction
Membrane electrode assembly (MEA) lifetime of greater than 40,000 hours remains an elusive goal of the fuel cell industry. A primary reason is the lack of fundamental understanding on the mechanisms of MEA degradation. Specifically, the relationship between component physical property changes and MEA performance decay has not been established. We report preliminary data relating changes in gas diffusion layer (GDL) physical properties to fuel cell performance decay.

Fuel cell performance decay can be separated into several separate mechanisms, one of which is changes in GDL surface properties as a function of oxidation. As a fuel cell runs, the GDL is exposed to an environment in which both chemical (hydrogen peroxide) and electrochemical (voltage) oxidation can occur. The GDL oxidation changes the hydrophobicity of the GDL, causing the MEA to flood by increasing the gas mass transport resistance through the GDL. Polarization curves taken over the course of a long-term fuel cell test shown in Figure 1 clearly illustrate the increase in mass transport resistance over time. It is the goal of this paper to show that GDL oxidation leading to decreased hydrophobicity is a probable cause for this type of performance decay. In all figures, the MEAs are made from dispersed Pt catalyst and PFSA ionomer. The MEA in Figure 1 uses a baseline GDL similar to GDL A in Figures 2 to 4 and Figure 6.

Figure 1. MEA performance decay over time. Test conditions: H₂/air at 800/1800 sccm; 70°C cell; anode and cathode inlet at 100% RH.
GDL Development
In order to address the problem of GDL oxidation, 3M developed test methods to measure GDL oxidative stability thereby focusing GDL developments. The first method involved submerging a GDL in a bath of acidified 15%wt H$_2$O$_2$ @ 180°F for a period of time. During this test, weight loss and the GDL static contact angle were monitored. Figure 2 illustrates the mass loss of the micro-layer of three different GDL designs after exposure to a peroxide bath. GDL C exhibited a slower mass rate loss than the other designs. The mass loss is attributed to complete oxidation of the carbon in the micro-layer.

![Figure 2. Mass loss of GDL micro-layer.](image)

The second test method involved applying a voltage to the GDL while in contact with an electrolyte. In this test, a corrosion current density is observed as a function of voltage. The corrosion current, or rate of oxidation, of GDL C at voltages greater than 1.5V vs. standard hydrogen electrode (SHE) was up to three orders of magnitude lower than GDL A or B, as shown in Figure 3.

![Figure 3. Corrosion current densities for different GDL designs.](image)
Accelerated Test Results

GDL oxidation is one mechanism for MEA performance decay. The methods of measuring GDL oxidative stability can be used to accelerate GDL oxidation. GDLs that were aged in hydrogen peroxide (test method #1) were then built into an MEA. A virgin GDL was placed on the anode and an aged GDL was placed on the cathode. Polarization curves were recorded as a function of time in the peroxide bath. The results for GDL A are shown in Figure 4. The results clearly indicate that GDL oxidation will reduce the MEA performance. The MEA performance loss seen in Figure 4 closely resembles the MEA performance decay seen in Figure 1 indicating that the peroxide accelerated aging conditions are representative of real life conditions. GDL C, which is more oxidatively stable, did not undergo the same type of performance decay. For GDL C, the polarization curves did not shift to the left, suggesting that the mass transport difficulties experienced by GDL A did not occur for GDL C (Figure 5).

![Figure 4. GDL A: Performance decay of an MEA with hydrogen peroxide aged cathode GDL. Test conditions: H₂/air at 800/1800 sccm; 70°C cell; anode and cathode inlet at 100% RH.](image)

![Figure 5. GDL C: Performance decay of an MEA with hydrogen peroxide aged cathode GDL. Test conditions: H₂/air at 800/1800 sccm; 70°C cell; anode and cathode inlet at 100% RH.](image)
The static contact angle of the aged GDL parallels the same trends seen in Figures 4 and 5. GDL A, which readily oxidizes, cannot maintain its original static contact angle. As the contact angle decreased, the performance at high current densities also decreased. On the other hand, the contact angle of GDL C remains quite stable (Figure 6).

Figure 6. Static contact angle as a function of time in hydrogen peroxide.

Conclusion
In order to meet the MEA lifetime goal of greater than 40,000 hours, methods of measuring and accelerating MEA decay mechanisms need to be completely understood; specifically, the relationship between component physical property changes and MEA performance decay. 3M has developed a method of screening new GDL designs as well as accelerating the oxidation of GDLs. With this tool set, a key link between the static contact angle and mass transport changes has been established and more stable GDLs have been developed.

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