## Control of Ionomer Distribution and Porosity in Roll-to-Roll Coated Fuel Cell Catalyst Layers

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Fuel cell catalyst layers are a complex mixture of catalyst particles and ion-conducting polymers. The performance of catalyst layers is highly dependent on microstructure, which must balance proton and electron conduction with gas transport. As fuel cells transition from the laboratory to industrial production for vehicles and other applications, there is a need to understand how manufacturing processes impact the microstructure and, ultimately, the performance of catalyst layers.

In this work, we have focused on the roll-to-roll coating of catalyst layers for proton exchange membrane fuel cells. The catalyst layers are responsible for the electrochemical reactions that enable power production. In these devices the catalyst is commonly a Pt nanoparticle supported on porous carbon blacks. The ion conducting polymer, or ionomer, is typically a perfluorosulfonic acid, the most common being Nafion. To fabricate the catalyst layer, the catalyst particles are dispersed in an ink with the ionomer and cast to form the catalyst layer. Commonly, laboratory samples are prepared using spray coating or hand painting, which effectively coat many thin layers to achieve the desired catalyst layer thickness. This creates catalyst layers with uniform distributions of materials throughout the thickness of the full catalyst layer.

In contrast, roll-to-roll methods, like slot die, gravure or screen printing, coat the catalyst layer as a single wet film. As the film dries, ink constituents are able to segregate, leading to a heterogeneous distribution of materials through the thickness of the layer. It is also known that the choice of solvent and drying rate can affect the morphology of the catalyst layer. Thus, there is a need to understand how ink formulation and the drying process influence the distribution of materials and catalyst layer morphology. Here, we have explored the influence of solvent and

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drying rate on morphology of roll-to-roll coated catalyst layers. We show the development of the Kelvin probe method as a rapid screening technique for qualitative analysis of ionomer distribution. We also utilize nano X-ray computed tomography to visualize electrode structure and to quantify particle-size and pore-size distributions, thickness-dependent ionomer distribution, tortuosity, and effective transport properties. We find that solvent has a strong influence on ionomer distribution, with less of an effect on porosity. Conversely, drying temperature has a strong influence on porosity, but less influence on ionomer distribution. Finally, we utilize in situ fuel cell performance testing and other advanced diagnostics to quantify the impact of catalyst layer properties on fuel cell performance and demonstrate that roll-to-roll coating is capable of coating high performance catalyst layers in multi-meter lengths.