Rheological Investigation of Catalyst Inks for Roll-to-Roll Processing of Fuel Cell Electrodes

Sunilkumar Khandavalli^a, Jae Hyung Park^c, Nancy N. Kariuki^c, Deborah J. Myers^c, Jonathan J. Stickel^b, Katherine Hurst^a, K. C. Neyerlin^a, Michael Ulsh^a, and Scott A. Mauger^a

^aNational Renewable Energy Laboratory, Chemistry Nanoscience Department, 15013 Denver West Parkway, Golden, CO 80401, USA

^bNational Bioenergy Center, National Renewable Energy Laboratory, Golden, CO 80401, USA ^cChemical Sciences and Engineering Division, Argonne National Laboratory Argonne, IL 60439, USA

ISCST- 20180918PM-B-PC4

Presented at the 19th International Coating Science and Technology Symposium, September 16-19, 2018, Long Beach, CA, USA[†].

Extended Abstract

Proton exchange membrane fuel cells (PEMFCs) are one of the promising energy technologies due to their clean energy emissions, high energy conversion efficiency for many applications as portable or stationary power sources and automobiles. The catalyst layer of a fuel cell membrane electrode assembly forms a critical component of a fuel cell. The microstructure of the CL controls the transport properties of electrons, protons, reactants, and products in the CL, which are key for fuel cell performance. One of the current challenges is the lack of rational approach to efficiently design the CL with an optimal microstructure for fuel cell performance [1].

The catalyst layer is commonly fabricated from an ink formulation through various deposition methods, predominantly lab-scale processes such as spray coating or hand painting. The microstructure of the catalyst ink formulation and how it is processed through coating and drying, strongly impacts the final catalyst layer morphology and the performance of fuel cell [2]. There has been growing interest in fabricating a catalyst layer through roll-to-roll coating methods due to a low-cost advantage. The rheological behavior plays a critical role in the processing behavior of the inks during the fabrication of electrodes and the evolution of final catalyst microstructure. To efficiently fabricate a catalyst layer with an optimal performance, an understanding of the structure-property-processing-morphology relationship is critical. Towards this goal, we conduct a rheological investigation on the microstructure of catalyst inks.

The catalyst ink system in this study consists of a mixture of platinum (47 wt%) loaded carbon black particles and ionomer (Nafion D2020) dispersed in water-alcohol (1-propanol, 1:3 mass ratio) solvent mixture. Two carbon supports used in the study were, a high surface area carbon (HSC, 800

[†] Unpublished. ISCST shall not be responsible for statements or opinions contained in papers or printed in its publications.

m²/g) and a low surface area carbon (Vulcan, 240 m²/g). We examine the effects of Pt catalyst particles loading on carbon support, ionomer concentration and carbon-support surface area on the rheological behavior of the catalyst ink dispersions in combination with dynamic light scattering (DLS) and ultra-small angle x-ray scattering (USAXS) experiments.

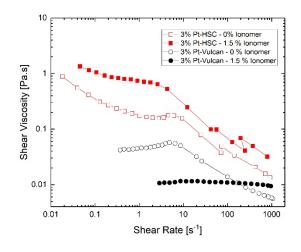


Figure 1. Steady shear rheology of catalyst inks comparing the effect of ionomer between inks with different surface area of the carbon support.

The neat carbon dispersions without ionomer or Pt present a strong shear thinning behavior indicating a highly agglomerated structure of carbon dispersions that is broken down by the shear flow fields [3]. The rheological behavior is sensitive to carbon-support surface area, where the low shear viscosity and the degree of shear thinning increased with the carbon-support surface area. This is due to a higher internal porosity and the agglomeration degree of a high surface area carbon compared to a low surface area carbon. The addition of Pt to the carbon particles resulted in a decrease in the low shear viscosity and the magnitude of shear thinning suggesting an electrostatic stabilization of the carbon aggregates by the charge of Pt and the carbon-support surface. The addition of the ionomer to the carbon dispersions with no Pt also results in a decrease in the viscosity. This is due to stabilization of carbon aggregates by the adsorbed ionomer through electro-steric mechanism. Interestingly, the addition of ionomer to Pt loaded carbon dispersions presented different rheological trends depending on the carbonsupport type: a low surface area carbon dispersion showed a decrease in the viscosity, whereas a high surface area carbon demonstrated an increase in the viscosity as shown in Fig. 1. The contrasting trends suggest strong differences in the ionomer distribution on the carbon surface based on its porous structure and the associated Pt distribution, which could strongly modify the inter-particle interactions and the bulk microstructure. The rheological observations were further verified using dynamic light scattering and ultrasmall angle x-ray scattering experiments.

References:

- [1] Holdcroft, S. Chem. Mater. 2014, 26, 381-393.
- [2] Hatzell, K. B.; Dixit, M. B.; Berlinger, S. A.; Weber, A. Z. J. Mater. Chem. A 2017, 5, 20527-20533.
- [3] Negi, S. A.; Osuji, O. C; *Rheol. Acta* 2009, 48, 871-881.