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Structure-to-Property Relationships in Non-Platinum Group Fuel Cell Catalysts: Examination of Length Scale Correlations

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Hydrogen fuel cells provide a means for direct electrical generation without the need for a mechanical-work intermediate. They are efficient and produce only water as a product [1]. Because they produce no carbon waste, fuel cells are means of green energy generation that are of great interest as a transportation energy source.

Fuel cells require catalysis to oxidize hydrogen and reduce oxygen at the anode and cathode, respectively. Current fuel cells rely on platinum as a catalyst, which is expensive and is a fundamentally limited resource [1,2]. Because of this, there is a significant amount of research being directed toward development of non-platinum group metal based catalysts. Nanostructured iron-nitrogen-carbon materials are active catalysts that have the potential to replace platinum as the cathode catalyst. However, targeted research on these materials is limited by a lack of understanding of how the physical structure and chemical composition of catalysts fundamentally contribute to their overall performance.

Catalyst performance is not dependent simply on chemical species or total surface area. There is a complex interplay between concentrations of different chemical moieties and morphology on different length scales. While the fact that the chemistry plays a crucial role seems self-evident, the roles of morphology are less straight-forward. Pore size and surface roughness length scales determine the efficiency and types of diffusion that allow fuel to be delivered to the catalyst surface and for removal of products [3]. Surface feature size is also related to how well water interacts with the surface (wettability) [4] and may play a role in the active sites themselves [5].

This presentation focuses on elucidation of catalyst morphology on different length scales. Surface morphology is explored using digital image processing (DIP) of SEM and AFM images. Previous DIP work has used high and low pass filtering to separate images by length scales [6–8]. Statistical measures of the decomposed images were then calculated and used for correlations to macroscopic measurements. However, this filtering method has not allowed for precise separation of length scales. It has not been possible to consistently separate microscale from mesoscale features.

Refinement of filtering techniques has yielded specific correlations between morphology and macroscopic properties across a range of Fe-N-C catalysts. The sample set shown in Figures 1 and 2 includes catalysts prepared from small C-N molecules and C-N polymers pyrolyzed with Fe salts, as well as pyrolyzed Fe containing macrocycles. There is a strong positive correlation between Fe-N center concentration and roughness on the scale of 15-300 nm. Half-wave potential, which is a measure of catalytic activity, is seen to increase as the contrast increases in filtered images containing features from 400-3000 nm.

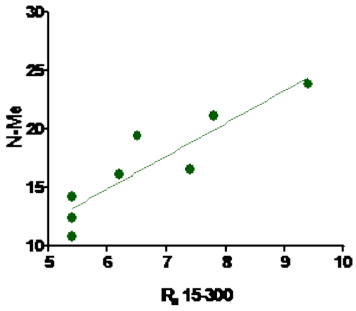

Figure 1: Relative concentration of Fe-N centers as measured by XPS vs. roughness on the scale of 15-300 nm.

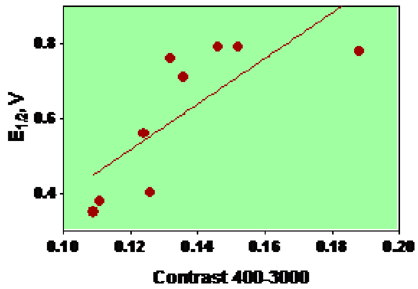

Figure 2: Half-wave potential measured by RDE vs. contrast on the scale of 400-3000 nm.

Current work explores the use of wavelet analysis on SEM images. Wavelet theory is a sub-branch of Fourier analysis. Unlike Fourier, which assumes an infinite periodic structure, wavelets are discreet. These individual, scalable wave shapes are fit to SEM images to extract new statistical information with finer length-scale separation than previous methods. These data are correlated with macroscopic surface area from BET, electrochemical performance from RDE, chemical speciation from XPS, and pore size volume and distribution of hydrophilic pores by water adsorption isotherms.

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