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Rotating Ring-Disk Study of Metal-Nitrogen-Carbon Catalyst Prepared by High Pressure Pyrolysis

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The need for less expensive oxygen reduction (ORR) catalysts has impeded commercialization of low temperature fuel cells for transportation applications. Metal/nitrogen/carbon (MNC) electrocatalysts represent promising non-precious materials that catalyze ORR (1-3). Preparation of MNC electrocatalysts involves the combination of metal and nitrogen components immobilized in a conductive carbon matrix by the pyrolysis of precursor materials. In the present work, the kinetics of a MNC catalyst was characterized with rotating ring-disk electrode (RRDE) studies. The results were analyzed with existing kinetic models.

Electrochemical characterization was conducted using a glassy carbon rotating ring-disk electrode. The rotating ring-disk method allows for controlled convection to the catalyst surface. The controlled convection allows a comparison of catalyst kinetics on the disk under a variety of conditions, while the platinum ring can be poised at a potential that detects stable intermediates (in this case peroxide) via a second electrochemical reaction. The study consists of steady-state current measurements at various potentials, rotation speeds, and catalyst loading. Furthermore, the experimental set is analyzed in terms of other aspects of the catalyst such as porosity, catalyst layer thickness, and peroxide generation.

Figure 1 shows polarization curves at various loadings. One can see that between 0.3 to 0.5 mg/cm^2 the polarizations curves and peroxide generation curves show relatively little change. But, below 0.3 mg/cm^2 the curves show significant loading effects. This suggests that increasing loadings above 0.3 mg/cm^2 does not impact oxygen utilization.

Below 0.8 V vs. RHE, where the ring current becomes significant, percent peroxide decreases with loading as shown in Figure 2. Although the disk current increases significantly with potential, the percent peroxide is primarily loading dependent with much less potential dependence. This indicates that although loadings above 0.3 mg/cm^2 may have little to do with oxygen utilization, they may be significant with regards to peroxide reduction.

For the considered MNC catalyst, over 95% of oxygen is fully reduced to water. Although the data suggests that peroxide reduction may have significant kinetic limitations, increased loading can improve performance, not only by improving oxygen reduction kinetics, but also by increasing electron efficiency through peroxide reduction.

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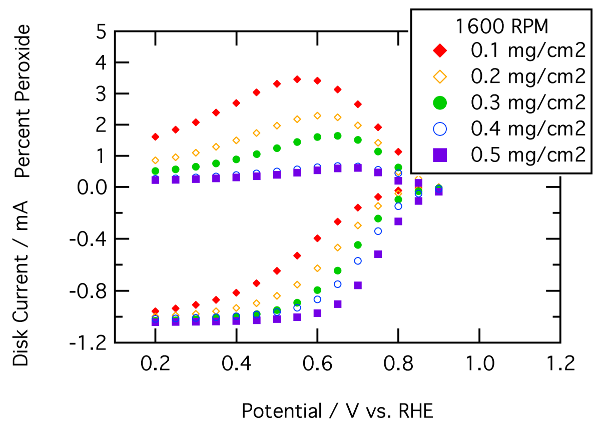

Figure 1. Steady-state oxygen reduction polarization, in 0.5 M H_2SO_4, room temperature, O_2-saturated, ring poised at 1.2 V vs. RHE at 1600 rpm with 5 mm diameter disk.

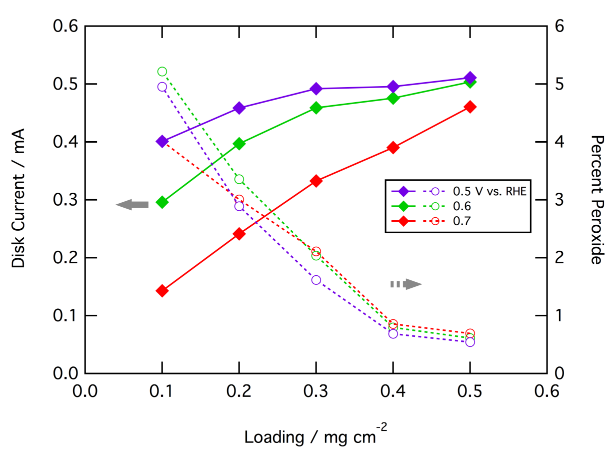

Figure 2. Disk current and percent peroxide vs. loading at various potentials: 400 RPM 0.5 M H_2SO_4, room temperature, oxygen saturated, ring poised at 1.2 V vs. RHE.