Pyrolysis Pressure Dependence of MNC Oxygen Reduction Electrocatalysts

Nathaniel Leonard, Selvarani Ganesan, Scott Calabrese Barton

Michigan State University
East Lansing, MI, 48824

Metal/nitrogen/carbon (MNC) electrocatalysts facilitate the oxygen reduction reaction (ORR) for low temperature fuel cells and may offer a replacement for precious metal electrocatalysts such as platinum. These compounds involve the combination of metal and nitrogen components immobilized in a conductive carbon matrix. The present work considers one method of producing these catalysts: pyrolysis of precursors in a closed vessel to contain gaseous intermediates and byproducts.

Pyrolysis is conducted in a sealed ampule containing metal, nitrogen, and carbon precursors. Here we study the effect of autogenic pressure generation on catalyst structure, morphology, and activity. Autogenic pressure is controlled by varying the specific mass of volatile precursors relative to the reactor volume [1-3].

The resulting catalysts were characterized electrochemically by rotating disk measurements and by polarization of membrane electrode assemblies (MEAs). The catalysts were compared based on current at 0.8 V vs RHE. For the precursor melamine using RDE studies, optimal nitrogen content was found at 25 wt.% nominal nitrogen, as shown in Figure 1. Lower nitrogen loading leads to lower final active site content, while excessive loading may decrease catalyst surface are via carbon deposition[1]. An ampule loading study was performed with the optimal mixture indicating that higher ampule loadings improve performance (Figure 2). We attribute this effect to increased gas-phase nitrogen activity driving increased active site density. Furthermore, this trend suggests that the limiting reaction may be an adsorption process, driven by high partial pressure of volatile intermediates.

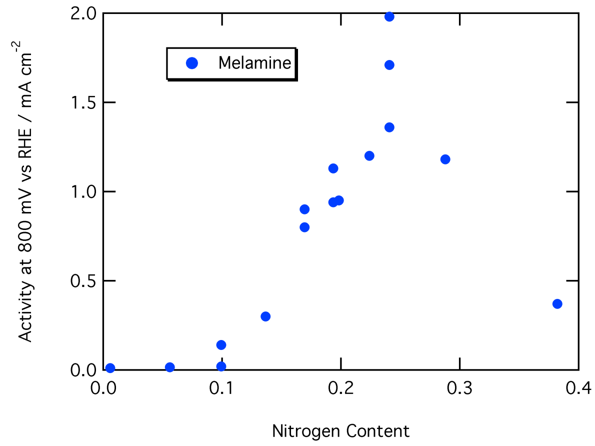

Figure 1. Nitrogen Optimization Study: constant mass to ampule volume loading

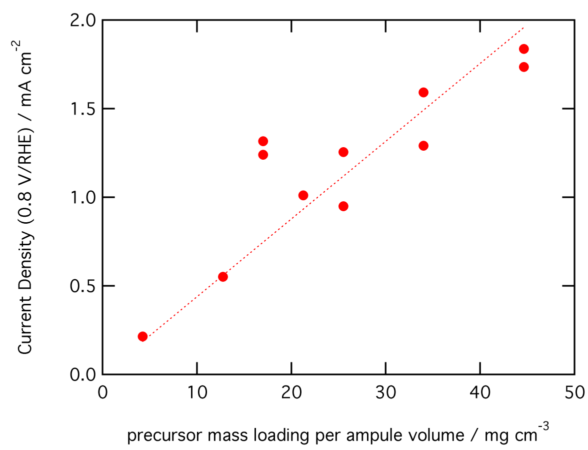

Figure 2. Ampule loading study: constant nitrogen:carbon:iron ratio.

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