Electrochemical Evidence for Two Sub-families of FeN_xC_y Moieties with Concentration-dependent Cyanide Poisoning

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Iron-nitrogen-carbon (Fe-N-C) catalysts are one of the most promising family of materials for the successful replacement of noble Pt in low temperature fuel cells. Despite recent advances in the synthesis, activity and site-structure of Fe-N-C catalysts, further improved understanding of the nature of the Fe-based active sites is still needed. In the present study, the existence of two sub-families of FeN_xC_y moieties is revealed by the concentration-dependence of cyanide poisoning on a Fe-N-C catalyst only comprising atomically dispersed iron. The analysis of the activity decrease upon contact with various cyanide concentrations and activity recovery following rinsing with water reveals the presence of two sub-groups of ORR-active FeN_xC_y species. They are discriminated by the reversibility or irreversibility of the cyanide poisoning. From Mössbauer spectroscopy study and poisoning test on a model compound Fe(II) phthalocyanine (FePc), we hypothesize that the reversibility/irreversibility of cyanide poisoning is governed by the competitive adsorption of oxygen molecule and cyanide anion.