Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

Electronic states of carbon alloy catalysts and nitrogen substituent effects on catalytic activity TOMOYUKI HATA, HIROSHI USHIYAMA, KOICHI YAMASHITA, Department of Chemical System Engineering, School of Engineering, The University of Tokyo — In recent years, Carbon Alloy Catalysts (CACs) are attracting attention as a candidate for non-platinum-based cathode catalysts in fuel cells. Oxygen reduction reactions at the cathode are divided into two elementary processes, electron transfer and oxygen adsorption. The electron transfer reaction is the rate-determining, and by comparison of energy levels, catalytic activity can be evaluated quantitatively. On the other hand, to begin with, adsorption mechanism is obscure. The purpose of this study is to understand the effect of nitrogen substitution and oxygen adsorption mechanism, by first-principle electronic structure calculations for nitrogen substituted models. To reproduce the elementary processes of oxygen adsorption, we assumed that the initial structures are formed based on the Pauling model, a CACs model and nitrogen substituted CACs models in which various points are replaced with nitrogen. When we try to focus only on the DOS peaks of oxygen, in some substituted model that has high adsorption activity, a characteristic partial occupancy state was found. We conclude that this state will affect the adsorption activity, and discuss on why partially occupied states appear with simplification by using an orbital correlation diagram.

Tomoyuki Hata
Department of Chemical System Engineering,
School of Engineering, The University of Tokyo

Date submitted: 27 Nov 2012 Electronic form version 1.4