主　論　文　要　旨

Study on Relationship between Electrocatalytic Activity and d-band Center of Pt Nanoparticles and New Electrocatalyst for Fuel cells

ふりがな　　とよだ　えいしろう
氏名　　Toyoda　　Eishiro

Polymer electrolyte fuel cells (PEMFCs) are one of the most promising next generation power source. For their wider commercialization, their cost and platinum (Pt) usage as electrocatalyst should be reduced. To achieve Pt reduction, we are aiming to clarify the relationship between the electrocatalitic (oxygen reduction reaction) activity and electronic structure, as a guide toward higher activity electrocatalysts.

In this study, validity of the relationship between the center of Pt5d band (d-band center) and activity, which is known for Pt-based bulk material, is examined for Pt nanoparticles, which are the present standard catalyst material. Pt nanoparticles with various sizes were deposited on the glassy carbon, and the activity and electronic structure were measured by rotating disk electrode method and synchrotron radiation photoelectron spectroscopy (SR-PES), respectively. Decreasing the particle size was found to lead to activity decrease and d-band center approach toward the Fermi level (E_F). This is consistent with the bulk Pt results showing that the closer the d-band center is to E_F, the lower the activity is.

To achieve a bulk Pt-like specific activity with a Pt thin layer, TiB_2(0001) was chosen as a substrate, because it is expected to have a strong affinity with Pt according to our DFT calculation. The activity of 4 monolayer (ML) Pt vacuum-deposited on TiB_2(0001) showed a similar activity with bulk Pt, although the catalyst showed rapid degradation. CO annealing, which is effective to obtain a uniform and flat surface on bulk Pt, was applied for 5 or 10 ML Pt on TiB_2(0001). These samples showed an activity equal to or higher than bulk Pt and much higher stability than the untreated thin Pt layers. This activity and stability are probably because the Pt thin layer is crystallized and alloyed with Ti.