Ab-initio Evaluation of Carbon Functionalized Pt/TiO₂ Catalyst for PEMFCs: A Interface Behavior

Jialong Li 1,2, Zhenbo Wang 1,2, Sue Hao 2, Geping Yin 1
1School of Chemical Engineering and Technology, Harbin Institute of Technology
2School of Science, Harbin Institute of Technology
Harbin, 150001 China, wangzhb@hit.edu.cn

As the most promising electrochemical energy conversion, fuel cells have been aware of its excellent efficiency, low carbon saving, and environmental friendly(1), among which the proton exchange membrane fuel cells (PEMFCs) attract extensively attentions for its excellent durability, low carbon saving, and environmental conversion, fuel cells have been aware of its excellent performance in durability of precious metal catalyst raise the cost of PEMFCs severely and deteriorate the share of electrical power market(4). Corrosion of carbon support and migration of noble metal particles which caused by extremely electrochemical circumstance are considered to be the main reason for performance degradation(5). Admittedly, alternative support materials, transition metal oxides(6, 7) and carbides(8), generate the improvement in durability, but activity absence still blocks the commercialization for insufficiency in electronic transport.

Titanium dioxide supported Pt catalyst has attracted considerable attention due to its excellent stability under acidic and oxidative circumstance recently. Huang(7) et al. reported the Pt/TiO₂ catalyst with ultrahigh stability, however, the initial activity is quintile of carbon support catalyst for the low electronic transport ability of titanium dioxide material. Hence, promoting the electronic conductivity is of importance. Modification of doping is the conventional route to modify the properties of transition metal oxides. However, few doping research is focusing on the interfacial behavior of catalyst support, which is crucial to the heterogeneous catalysis. It is illustrated that the interfacial functionalized process can enhance the activity of titanium dioxide supported Pt catalyst.

The computer simulation technique provides effective solution to investigate the electronic behavior on the micro scale of interface, which is difficult for in situ analysis. With the progress in computing capacity and algorithm, the ab-initio evaluation can propose reliable theoretical prospect for elevating the performance of material. In this work, the periodic density functional theory (DFT) calculations were performed with CASTEP code and hybrid DFT method (9, 10) to elevated the electronic behavior during the interaction between Pt cluster and carbon functionalized TiO₂ interface which is barely discussed. The model system of Pt nanoparticles were described as an optimized Pt cluster (6 Pt atoms) supported on (4×4) supercell anatase TiO₂ (101) interface. The interaction energy between Pt cluster and TiO₂ interface was obtained as follow equation

$$E_{\text{inter}} = E_{\text{cluster+interface}} - E_{\text{cluster}} - E_{\text{interface}}$$

where $E_{\text{cluster+interface}}$ is the total energy of the interaction system of TiO₂ supported Pt cluster, and $E_{\text{cluster}}$ is the total energy of optimized Pt cluster, and $E_{\text{interface}}$ is the relax optimized bare anatase TiO₂ (101) interface. Thus, the $E_{\text{inter}}$ indicates the interaction intensity between the cluster and interface. The finally ab-initio evaluation illustrated that the carbon interfacial functionalized process significantly promotes the interaction intensity between TiO₂ and Pt nanoparticles, which proposes the probably solution for reconcile of stability against activity paradox.

![Fig. 1. The model system of carbon functionalized Pt/TiO₂ catalyst](image)

Reference