第一原理計算を用いたカーボンフリーPt ナノ粒子連結触媒の酸素還元活性向上要因の解明

(東京工業大学[1]・神奈川県立産業技術総合研究所[2]・高度情報科学技術研究機構[3])
○蒲田瑞季[1]・黒木秀記[1,2]・菅原勇貴[1]・牛山浩[3]・山口猛央[1]

Elucidation of factors for the enhanced oxygen reduction activity of carbon-free connected Pt catalysts using first-principles calculation

([1]*Tokyo Institute of Technology*, [2]*Kanagawa Institute of Industrial Science and Technology (KISTEC)*, [3]*Research Organization for Information Science and Technology (RIST)*) ○Mizuki Kamata,[1] Hidenori Kuroki,[1,2] Yuuki Sugawara,[1] Hiroshi Ushiyama,[3] Takeo Yamaguchi[1]

The development of highly active oxygen-reduction-reaction (ORR) catalysts is required to spread the use of polymer electrolyte fuel cells. We have developed carbon-free connected Pt-based catalysts (Pt capsule) comprising of nanonetworks formed by the connection of Pt-based nanoparticles (Fig. 1a).[1)] The connected Pt catalyst shows four times higher ORR specific activity than a conventional carbon-supported Pt nanoparticle catalyst (Pt/C); however, the factors in the enhanced activity are not yet clarified (Fig. 1b). In this study, we constructed various models that simulated the structures of a carbon-free connected Pt catalyst and Pt/C to evaluate the adsorption energies of ORR intermediates (oxygen and water) on Pt atoms using first-principles calculation. As a result, we found out that the presence of carbon support would affect the adsorption of oxygen on the Pt atoms, indicating that a carbon-free structure is an important factor for an enhanced ORR activity.

Keywords : First-principles Calculation; Oxygen Reduction Reaction; Fuel Cells

固体高分子形燃料電池の普及拡大のため、酸素還元反応(ORR)触媒の高活性化が求められている。本グループは、Pt 系ナノ粒子が連結したナノネットワークで構成されるカーボンフリーPt 系ナノ粒子連結触媒(Pt capsule)を開発している(図 1a)[1)]。Pt ナノ粒子連結触媒は、従来の Pt ナノ粒子担持カーボン(Pt/C)より 4 倍高い ORR 表面比活性を示すが、その要因は明らかとなっていない(図 1b)。そこで、本研究ではカーボンフリーPt ナノ粒子連結触媒と Pt/C の触媒構造を模擬した様々なモデルを構築し、Pt 上での ORR 中間体(酸素や水)の吸着エネルギーを第一原理計算により評価した。その結果、カーボン担体の存在が Pt 上への酸素種吸着エネルギーに影響することが分かり、カーボンフリー構造は ORR 高活性化に重要であることが示唆された。

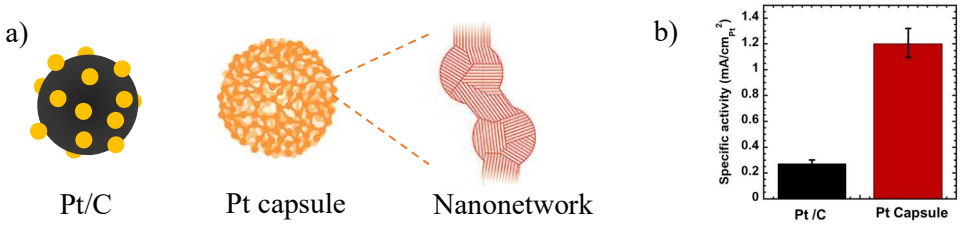

図 1 a) Schematics of Pt/C and Pt capsule catalysts, and b) ORR activity of the catalysts

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1) T. Yamaguchi, *et al.*, *Energy Environ. Sci.*, 2015, **8**, 3545–3549.