

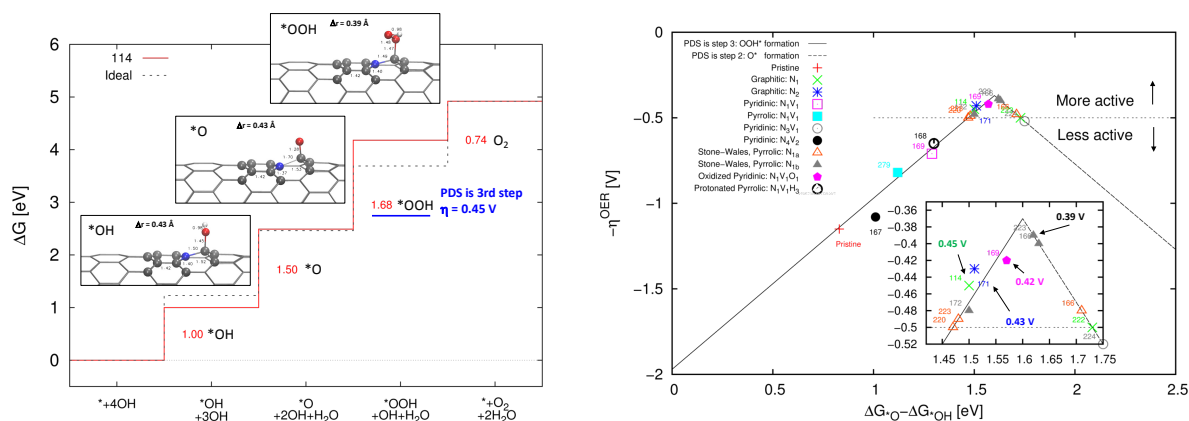
Oxygen evolution reaction on nitrogen-doped carbon nanotubes

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A renewable energy ecosystem requires an energy carrier such as hydrogen obtained by electrochemical water splitting to store the often intermittent energy generated from carbon-free sources. The ideal catalyst material should be efficient, stable under operating conditions, and composed of abundant elements. Density functional theory simulations within a thermodynamic model [2] of the more difficult half-reaction, the anodic oxygen evolution reaction (OER), with a carbon nanotube as catalyst, show that the presence of $< 1\%$ nitrogen reduces the required OER overpotential significantly. Various nitrogen functionalities (graphitic, pyridinic, Stone-Wales pyrrolic, oxidized) result in model catalysts with predicted overpotentials of 0.39–0.45 V, which is near the peak of the volcano plot in this model and close to what has been observed experimentally for nitrogen-doped nanotubes [1]. The nanotube radius and chirality effects are examined by considering the analogous graphene systems which yield similar trends but with reduced reactivity. For the majority of the systems, the third step of the four step OER mechanism, the formation of attached OOH, is the rate-determining step [3]. Our study of kinetic barriers will also be briefly discussed [4].



Left: Thermodynamic free energy step plot for OER on the graphitically N₁ nitrogen-doped single-walled carbon nanotube (SWNT). Right: Volcano plot for OER on SWNT-based model systems. The inset highlights the best performing systems.

References

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