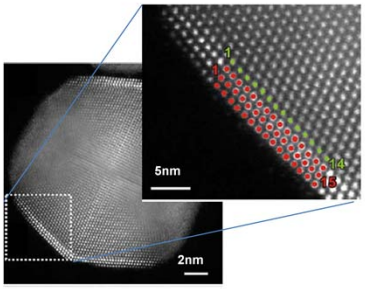
Quadrupling the Activity of Core-Shell Catalyst Particles



Transmission electron microscopy imagaes showing the morphology and atomic structure of typical Pd@$Pt_{2.7L}$ icosahedra nanoparticles

Work was performed as a User Project at the Center for Nanophase Materials Sciences, Georgia Institute of Technology, University of Wisconsin-Madison, and Xiamen University.

Scientific Achievement

Electron microscopy reveals that lattice distortions in Pd-Pt core-shell catalyst particles lead to four- to eight-fold enhancements in activity for oxygen reduction.

Significance and Impact

This discovery leads to a novel and effective synthesis approach for electrocatalysts with greatly enhanced activity and durability.

Research Details

- High-resolution STEM imaging revealed unique atomic structures with coexisting twin boundaries and strained core-shell arrangements, resulting in lateral confinement and compressive strains within Pt surface layers.
- Specific and mass activities toward the oxygen reduction ORR are enhanced 8X relative to a commercial Pd@Pt catalyst and after 10,000 cycles, mass activity is 4X higher than the commercial catalyst.

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Narrative Highlight Text:
Catalytic activities of bi-metallic catalysts significantly relay on their detailed atomic structure and elemental distribution at surface or near-surface region. Here we report design of novel icosahedra structured Pt@Pd nanoparticles. In addition to the icosahedra morphology, electron microscopy reveals its unique atomic structures of such particles: the coexistence of twin boundaries and strained core-shell structure. Owing to lateral confinement imposed by twin boundaries and thus vertical relaxation only, the platinum overlayers evolve into a corrugated structure with compressive strains. For the core-shell nanocrystals with 2.7 platinum overlayers, their specific and mass activities toward oxygen reduction are enhanced by eight and seven folds, respectively, relative to a commercial catalyst. Density functional theory calculations indicate that the enhancement can be attributed to the weakened binding of hydroxyl to a compressed platinum surface supported on palladium. After 10,000 testing cycles, the mass activity of the core-shell nanocrystals is still four times higher than the commercial catalyst. These results demonstrate an effective approach to the development of electrocatalysts with greatly enhanced activity and durability.

Reference
"Palladium-platinum core-shell icosahedra with substantially enhanced activity and durability toward oxygen reduction"
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