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Title:	Shape- controlled synthesis of palladium based nanostrucyures for electrochemical energy harvesting and organic transformation reactions
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Abstract:	Nanomaterials have gained enormous attention over the last century due to their immense importance in the field of catalysis. The development of various chemical industries and newenergy resources (e.g., lithium-ion batteries or fuel cells) have relied mostly on noble metal based nanomaterials. Among various noble metal nanostructures, Pd nanocrystals are of great interest owing to their extensive applications in diverse catalytic fields, such as electrocatalytic oxidation of small molecules including formic acid, methanol, and ethanol, or reduction reactions including oxygen reduction reaction (ORR), and hydrogen evolution reaction, and also in organic transformation reactions like C-C bond formation reactions (Suzuki or Heck coupling) and catalytic hydrogenation reactions. The versatile use of palladium catalyst for various applications can be attributed to its ability to stabilize different oxidations states. However, the electrocatalytic efficiency of Pd nanostructures is not at par with Pt-based nanocrystals, which is used in commercial devices. This provides intense motivation towards the development of Pd-based nanocrystals which can act as a potential replacement of scarce Pt catalysts in fuel cell applications. Additionally, Pd nanocrystals behaving as heterogeneous catalysts lack high reactivity as well as selectivity as compared to their homogeneous analogs. Moreover, catalyst recovery and recycling are crucial aspects considering both environmental point of view as well as industrial application purposes which is also energy demanding. Thus, there is plenty of room for the development of highly efficient Pd-based nanostructured catalysts for the above mentioned applications. This thesis describes the synthesis of efficient Pd-based nanostructures with various shapes such as nanocube (NCs), long nanowires (NWs), twisty nanowires (TNWs), nanoparticles (NPs), etc., and their application in electrocatalytic energy harvesting reactions as well as organic transformation reactions. To improve the efficiency of Pd NPs, I have deposited Pd atom-by-atom on an N doped reduced graphene oxide (NRGO) which exhibited very high ORR performance due to the presence of strong interaction between Pd and pyrrolic-fraction of the N-moieties present in NRGO. Then, I have shown that Pd nanocubes containing highly stable (100) facets can be synthesized by facile one-pot synthesis route in water assisted by halide ions and poly(vinylpyrrolidone) under the precise control of acid etching which has a natural tendency to form a self-assembled two-dimensional superlattices in the absence of any additional chelating agents. These Pd NCs demonstrated superior bifunctional electrocatalytic behavior for the ORR as well as methanol oxidation reactions (MORs). Furthermore, I have shown that alloying Pd with Ni and controlling the morphology of the nanocrystals to generate ultrathin TNWs improves their electrocatalytic efficiency for ORR enormously. Importantly, these PdNi TNWs also exhibited extremely high stability up to 2,00,000 catalytic cycles with minimal loss in catalytic activity while maintaining an efficiency significantly higher than the DOE-USA target-2020 throughout. Since Pd is also an active catalyst for many organic transformation reaction, I have investigated the fate of Pd nanostructures for Suzuki copling reaction, and reduction reactions. I discovered that synthesizing Pd NCs on NRGO support by atom-by-atom addition of Pd lowers the d-band center of Pd and improves the charge transfer resistance, thereby exhibits very high catalytic efficiencies for C-C cross-coupling reaction and nitroarene reduction reactions. I will be also be discussing about improving catalytic efficiencies of the Pd nanocrystals for Suzuki- Miyaura cross-coupling and hydrogenation reactions by tuning the molecular diffusion near the catalyst-surface across the ligand-shell of polyvinyl-pyrrolidone. Furthermore, I found that these Pd nanostructures are highly compatible with various types of solids supports such as cellulose paper and polyurethane foam, permitting their immobilization which results in their efficient recycling, a foremost important aspect for developing sustainable industrial processes.
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