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PAPER

Structure and stability of platinum nanocrystals: from low-index to high-index facets

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High index surfaces are introduced into Pt nanocrystals because they are expected to exhibit higher catalytic activity than low index planes such as {111}, {100}, and even {110}. This article presents a systematic investigation on the structure and stability of polyhedral Pt nanocrystals with both low-index and high-index facets by means of atomistic simulations. It has been found that the stability of Pt nanocrystals depends strongly on the particle shape and surface structures. Those nanocrystals, enclosed by high-index facets of {310}, {311}, and {331}, possess better stability and higher dangling bond density of surface compared with those ones with low-index facets, such as {100} and {110}, suggesting that they should become preferential candidates for nanocatalysts. The octahedral nanocrystals with {111} facets, though they have excellent structural and thermal stabilities, present the lowest dangling bond density of surface.

1. Introduction

Platinum (Pt) is the unique catalyst for many important reactions applied in modern chemical industry, petrochemical industry, automobile exhaust purification, and fuel cells because of its excellent activity and stability.^{1,2} The price of Pt, however, is extremely high and its reserve is rare on the earth, limiting its further wide use. In order to meet the increasing demand in industry, to explore and design new types of Pt catalysts with enhanced activity and stability therefore becomes the key issue in development of industrial catalytic fields.

It is well-known that the exceptional chemical (especially the catalytic) and physical properties of nanocrystals (NCs) are determined not only by the large portion of surface atoms but also by the surface structures of the particles.³ The former is determined by the particle size, and the latter relies on the particle shape. Available studies have also demonstrated that the catalytic functionality of metal NCs was not only affected by their size but also highly related to their shapes depending on the crystallographic planes that terminate the NC surface.^{4,5} Therefore, the shape-controlled synthesis of NCs becomes a promising direction for precisely tuning the activity, stability, and selectivity of NC catalysts. After the pioneering work of El-Sayed *et al.* for the synthesis of cubic and tetrahedral Pt NCs,⁶ many efforts have been devoted to prepare Pt NCs with different shapes by changing Pt precursors, reducing reagents,

stabilizing reagents, and solvents. So far, there are experimentally synthesized Pt NCs catalysts with cube, tetrahedron, octahedron, cuboctahedron, decahedron and icosahedron, which are usually enclosed by low index facets of {100} and {111}.^{7–14} Fundamental studies on the single-crystal surfaces of bulk Pt have revealed that high-index planes exhibit much higher catalytic activity than these low-index planes, because high-index planes have a greater density of unsaturated atomic steps, ledges, and kinks which can serve as active sites for breaking chemical bonds.¹⁵ It is naturally expected that high index surfaces should be introduced to improve the catalytic activity of Pt NCs. This is, however, rather difficult owing to the thermodynamical morphology preference, which is decided by the crystal growth rules: NCs have a strong tendency to form into structures with a minimization of the total surface energy, *i.e.* the NCs are preferentially enclosed with {111} and {100} low-index facets on which the atoms are closely packed,¹¹ resulting in the rapid elimination of high-index planes during particle formation. A breakthrough progress has been achieved by Sun and co-workers who have successfully prepared tetrahedral Pt NCs bounded with {730} high-index facets by developing an electrochemical square-wave potential method to control the growth and the surface structure of Pt NCs.^{16–18} These unusual Pt NCs exhibited good stability and greatly enhanced catalytic activity compared with the existing commercial Pt catalysts.^{19–22} Their work has stimulated great interest in synthesis, characterization and theoretical investigation of those Pt NCs with high-index facets.

Despite the fact that the synthesis and catalytic properties of Pt NCs have been extensively investigated, to the best of our knowledge, a thorough study of surface structures and stabilities of polyhedral Pt NCs with different sizes and shapes is still

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Structure And Stability Trends In The Platinum Metalsilicon Systems

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