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学　位　論　文　内　容　の　要　旨

博士の専攻分野の名称　　博士（工学）　　氏名　DENG Lianlian

学　位　論　文　題　名

Sputter deposition and formation mechanism of Pt and Pt alloy nanoparticles
(スパッタリング法による Pt と Pt 系二元金属ナノ粒子生成と形成メカニズムの研究)

Platinum (Pt) and Pt alloy nanoparticles (NPs) have been the subject of intense study, especially in catalysis. Chemical reduction methods often use reducing agents and stabilizing agents to produce metal NPs, and hence, the NPs can contain byproducts which can influence the catalytic properties. In addition, the synthesis of alloy NPs can result in core-shell structure or phase segregation due to different reduction potentials of different metal ions. To solve these issues, synthesis of metal and metal alloy NPs by vacuum sputtering onto a liquid matrix is proposed. The method is a green approach because it neither uses toxic reducing agents, nor produces any by-products. Further the top-down vacuum technique can generate metal atoms/clusters from the bulk metal targets, regardless the different reduction potentials of different metals, making the synthesis of solid solution alloy NPs become feasible. A low volatile liquid medium can be used in vacuum sputtering to control the growth, disperse, and stabilize NPs. However, the stability, growth, and formation mechanism of NPs are still the subject of discussion and dependent on multiple sputtering parameters. Thus, this study aims to synthesize Pt and Pt alloy NPs with desired size and understand the stability, growth and formation mechanism of the synthesized NPs.

Synthesis of highly uniform Pt NPs with small size (below 2.0 nm) and narrow size distribution by sputtering of Pt onto polyethylene glycol (Mw. = 600) is the topic of chapter 2. The results indicate that particle sizes were tailorable from 0.9 ± 0.3 nm to 1.4 ± 0.3 nm by varying the sputtering current (5 – 50 mA) with negligible particle aggregation occurred in PEG during sputtering. It is found that a slight growth of particle size after sputtering can be attributed to the addition of free Pt atoms to the existing Pt NPs. All samples formed stable dispersions in PEG for 5 months storage. This result suggests an advantage of using a liquid matrix to produce and stabilize NPs.

Chapter 3 in the thesis is devoted for the synthesis of well-dispersed and stable Pt/Cu alloy NPs and studying growth mechanism of the alloy NPs in sputtering of Pt/Cu alloy target onto PEG. The effects of sputtering current, rotation speed of the stirrer, sputtering time, sputtering period, and temperature of PEG on the particle size are studied systematically. The key results demonstrate that the aggregation and growth of Pt/Cu alloy NPs occurred at the surface as well as inside the liquid polymer after the particles landed on the liquid surface. According to particle size analysis, a low sputtering current, high rotation speed for the stirrer, short sputtering period, and short sputtering time are found to be favorable for producing small-sized single crystalline alloy NPs. On the other hand, varying the temperature of the liquid PEG does not have any significant impact on the particle size. Thus, these findings shed light on controlling NP growth using the newly developed green sputtering deposition technique.

Chapter 4 targets to the synthesis and formation mechanism of Pt/Au alloy NPs in a wide composition

range by simultaneous sputter deposition of two independent magnetron sources onto PEG (Mw. = 600). The resulting NPs are alloy with the face-centered cubic (fcc) structure. It is observed that the particle sizes, composition, and shape are strongly correlated. On the other hand, these characteristics can be tailored by varying sputtering parameters appropriately. Large agglomerates are formed at Pt content less than 20 at. %, showing partial alloy structure. Highly dispersed NPs with no agglomeration in PEG can be obtained when Pt is more than 34 at. %. Moreover, a small amount of Pt could terminate the agglomeration of Au when sputtering on TEM grid. Experimental results for 30 min sputtering onto PEG for various sputtering currents, as well as computational simulations, indicate that using the formation energy and selective attachment during particle formation can be used for explaining the composition-dependent particle-size of Pt/Au NPs.
Finally, research findings, contribution, and perspective are summarized in chapter 5 of the thesis.