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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（理学） 氏名 魏 金建

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Dynamic Process of Gold Nanoparticle Assembly using Fluorinated Surface Ligands in Solutions

（溶液中におけるフッ素化リガンド分子修飾金ナノ粒子の動的な自己集合プロセスに関する研究）

Gold nanoparticle vesicle (GNV) with a hollow interior and uniquely collective chemical and physical properties, have attracted particular attention for their potential applications. But, the dynamic self-assembly process or the formation of GNVs in solution was limitedly understood. The fundamental understanding of the formation mechanism of GNVs in solution can enhance our ability to manipulate and simplify their potential applications.

In this thesis, two new types of small fluorinated surface ligands, glucose-terminated fluorinated-oligo(ethylene glycol) ligand (GFL) with a neutral head as a presentative sugar-terminated fluorinated-oligo(ethylene glycol) ligand and carboxylic acid-terminated fluorinated-OEG ligand (CFL) with a weak negatively charged head, were designed respectively. And the dynamical self-assembly with GNPs using these two types of ligands or formation of assemblies in solution were extensively investigated.

Chapter 1 starts from an overview of static and dynamic self-assembly of molecules. A brief summary of dynamic self-assembly of metal nanoparticles and the nanoscale forces involved in the self-assembly are further reviewed. This chapter inspires the further design of new types fluorinated ligands based on our previous study for the investigation on their controlled self-assembly.

Chapter 2 describes the formation of GNVs composed of 15 nm GNPs or GNV-like assembly composed of 30 nm GNPs using GFL in solution and the formation mechanism of GNV-like assembly. The concentration of Au-30 NPs can determine the size of GNV-like assembly. The dynamic self-assembly process of GFL and 30 nm GNPs was further extensively investigated by the time-dependent studies of UV-vis, dynamic light scattering (DLS) and electron microscopy measurements. These results supported that a quick aggregation and slow self-assembly process were involved in the formation of GNV-like assembly in solution, indicating that both kinetics and thermodynamics were involved in the dynamic self-assembly process to produce thermodynamically stable GNV-like assemblies with a saturated size. A snap shot obtained from X-ray laser diffraction imaging strongly supported that GNV-like assembly composed of 30 nm GNPs was rawly formed in solution rather than during a drying process.

Chapter 3 describes the formation of GNV-like assembly composed of 15 nm GNPs or 30

nm GNPs using a carboxylic acid-terminated fluorinated-OEG ligand (CFL) with weak negative charge and the formation mechanism study of GNV-like assembly. The concentration of Au-30 NPs showed independence on the size of the GNV-like assembly. Effect of NaCl on the self-assembly process of CFL and 30 nm GNPs revealed that an important influence of electrostatic repulsive interaction provided by the carboxylic acid head on the formation of GNV-like assembly. Time-dependent study of self-assembly process of CFL and Au-30 NPs using UV-vis, DLS and electron microscopy measurements supported that a kinetic process was involved in the formation of thermodynamically stable GNV-like assembly.

Chapter 4 describes the formation of yolk/shell assembly by the size-segregation of binary mixtures of GNPs in the presence of GFL or CFL in solution. Time-course studies revealed that the size segregation occurred at the early stage of the self-assembly. These kinetically trapped aggregations transformed to energetically stable size-segregated yolk/shell structure. A snap shot obtained from X-ray laser diffraction imaging clearly indicated that the size-segregated assembly was rawly formed in solution.

Chapter 5 summarizes the main results in each chapter.