

Self-assembly of low dimensional nanostructures and materials via supramolecular interactions at interfaces

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ABSTRACT

Low dimensional inorganic–organic heterostructure nanomaterials with multiple different functional components attaching at interface have drawn considerable attention over the last decades, due to their excellent optical and electronic properties in comparison with their individual components. Supramolecular self-assembly of disordered building blocks has played an essential role in constructing well-defined functionalized structures. Solid–liquid interfaces are known as ideal platforms in the self-assembly process, thus providing a series of novel and attractive strategies to construct low dimensional inorganic–organic heterostructure nanomaterials with different sizes and shapes. In this review, we summarized low dimensional inorganic–organic heterostructure nanomaterials on the basis of self-assembly that is driven by synergistically metal–ligand coordination and other intermolecular interactions. Both stepwise self-assembly on the inorganic (or organic) solid surface and self-assembly of inorganic–organic building block through hydrogen bond, metal–ligand coordination interaction, π – π stacking, hydrophobic interaction, and covalently bonding were discussed.

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1. Introduction

Low dimensional inorganic–organic heterostructure nanomaterials are an important class of functional materials that comprise two different components attaching at interfaces. Recently, fabrication of such heterostructure materials with defined functions, such as inorganic semiconductor junctions [1], metal–metal junctions [2], inorganic semiconductor–metal junctions [3], and inorganic–organic semiconductor junctions [4], has drawn increasing attention due to their potential applications in the area of nano-electronic and optoelectronic devices [5–10]. Low dimensional inorganic–organic heterostructure nanomaterials consist of functional organic molecules attached on the surfaces of inorganic components in a covalent or noncovalent manner. Integration of inorganic and organic materials can take advantages of both the excellent optoelectronic properties of the former and remarkably improved stability of the latter. Moreover, readily modification of the organic components might allow tailoring of the resulting chemical and optoelectronic properties. Particularly, the formation of aggregated nanostructures at the inorganic–organic interface often causes superior optical and electrical properties to those of their individual components, namely the so-called synergistic performance ($1 + 1 > 2$). The resulted inorganic–organic heterostructures have drawn increasing interests for their applications in the

field of chemosensors [11], photodetectors [8], light-emitting diodes (LEDs) [12], and field-effect transistors (FETs) [13].

Since the performance of functionalized inorganic–organic heterostructures is extensively dependent on molecular parking and orientation at the inorganic–organic interface, self-assembly of small building blocks has played an essential role in constructing well-defined functionalized structures of unexpected supramolecular arrangements and functionalities. Based on various specific noncovalent interactions, such as hydrogen bonding, electrostatic forces, hydrophobic interaction, and metal–ligand coordinations, self-assembly process can drive the disordered building blocks into well-defined patterns spontaneously. Rational design of inorganic–organic heterostructures with a variety of morphologies including knots, rings, cages, helicates, ladders, and grids in bulk systems has been increasingly reported over the last decades [14–17]. In addition to the specific noncovalent interactions, the assembled condition also has a significant impact on the structure and stability of the assembled systems. Two-phase interfaces such as solid–liquid interface, liquid–gas interface, or liquid–liquid interface undergo particular interfacial effects [18–21], thus is a well-established environment for self-assembly of building blocks, which is in many ways different from the self-assembly in bulk systems. The solid–liquid interfaces which include solid substrates provide the interfaces good mechanical stability, and these interfaces could exist in a variety of geometries and morphologies (particles, rods, tubes, etc.). Therefore, the solid–liquid interfaces could be considered as ideal platform in the self-assembly process, thus providing a series

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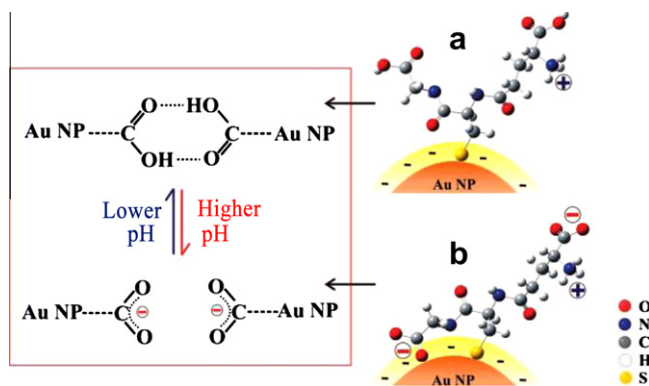


Fig. 1. Schematic illustrations of the pH-modulated interparticle interactions for assembly and disassembly of the GSH protecting AuNPs [32].

of novel and attractive strategies to construct inorganic–organic heterostructures with different sizes and shapes.

The strong and directional metal–ligand coordination interaction is proved to be an ideal driving force for attaching the inorganic and organic components together, thus can be conveniently used to construct inorganic–organic heterostructures [22,23]. Synergistic performance of other intermolecular interactions could lead to a wide range of aggregated morphology of the inorganic–organic heterostructures with different optical and electronic properties. To construct the self-assembled low dimensional inorganic–organic heterostructure nanomaterials at solid–liquid interface, there are two main strategies: ① fabricating solid inorganic (or organic) compositions that can be used to self-assemble organic (or inorganic) parts onto the solid surfaces; ② fabricating inorganic or organic building blocks that can be further readily self-assembled into well-defined structures. This review summarized low dimensional inorganic–organic heterostructure nanomaterials on the basis of self-assembly that is driven by synergistically metal–ligand coordination and other intermolecular interactions. In the first part of this review, self-assembly of inorganic–organic building block was discussed, especially taking Au nanoparticles (AuNPs) as example. This part was further expanded according to different driving forces during the self-assembly of the AuNPs, such as hydrogen bond, metal–ligand coordination interactions, π – π stacking, hydrophobic interactions, and covalently bonding. In the second part, stepwise self-assembly on the inorganic (or organ-

ic) solid surface was elaborated, to construct low dimensional inorganic–organic heterostructure nanomaterials.

2. Self-assembly of inorganic–organic building blocks

Recently, zero-dimensional inorganic nanoparticles (Au, GdSe, ZnO, etc.) are attracting increasing interest due to their excellent electronic, photonic, magnetic, and catalytic properties [24–26]. A large variety of strategies have been achieved for synthesizing stabilized inorganic nanoparticles with different sizes and shapes. It is known that nano-sized particles possess large surface areas, thus tending to aggregate and flocculate when dispersed in solvent. Upon addition of suitable organic ligand into the solvent, the self-assembly process at the solid–liquid interface would stabilize the dispersed nanoparticles, via metal–ligand coordination interactions. According to the compositions of inorganic nanoparticles, many different organic ligands have been designed and synthesized. For example, gold nanoparticles (AuNPs) are well known as a class of widely investigated nano-sized metal materials owing to their facile surface functionalization, chemical stability, size-related electronic, optical, and magnetic properties. Lots of typical ligands, such as thiols, xanthates, disulfides, amines, ammonium salts, carboxylates, and phosphines, have been employed for the self-assembly process at the AuNPs surface [27].

Obviously, the inorganic nanoparticles attached with organic ligands are exactly nano-sized inorganic–organic heterostructures. In a “bottom-up” manner, these nanoparticles with inorganic–organic hybrid nature can be used as building blocks for further self-assembly process, so as to construct well-defined structures. During the self-assembly of the nanoparticle building blocks, the driving forces are of paramount importance for achieving desired structures and functionalities. Therefore, rational and precise design could efficiently direct the self-assembly process of the nano-sized building blocks with inorganic–organic hybrid nature.

2.1. Self-assembly through hydrogen bonding

During the past decades, hydrogen bonding has drawn considerable attention for its applications in the field of functional material science and biochemistry, due to its directionality and specificity [28,29]. A large variety of well-defined functionalized aggregates have been successfully fabricated via hydrogen bonding, for developing novel nanoelectronic materials and devices.

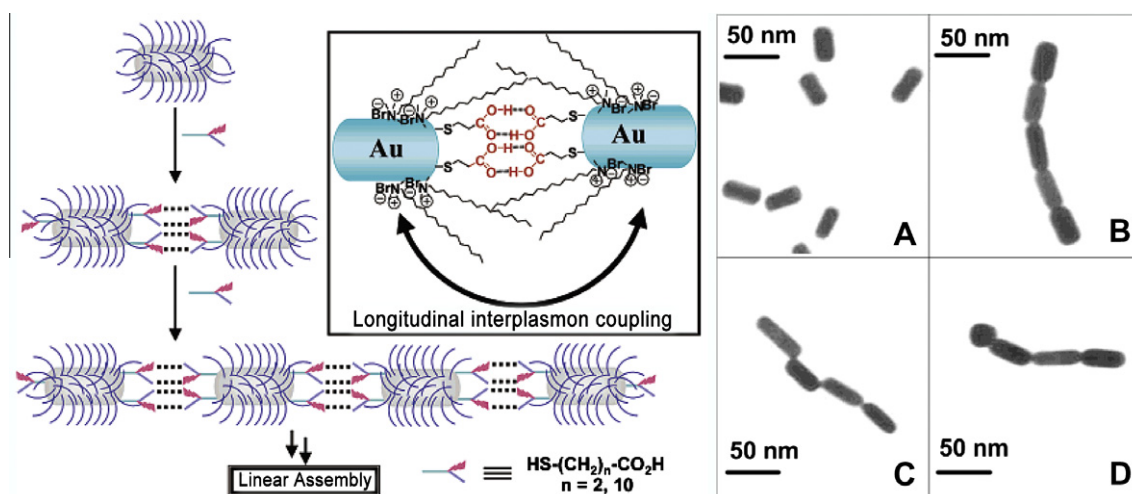


Fig. 2. Pictorial representation of the linear self-assembly of Au nanorods. TEM images of Au nanorods (A); three separate examples of the linear assembled aggregates of Au nanorods (B–D) [34].

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