



## Self-assembly of particles—The regulatory role of particle flexibility

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### ABSTRACT

Aspects of the self-assembly of particles, which uses nanometer or micrometer sized building blocks to bridge the gap between microscopic and macroscopic scales, are reviewed. Particle self-assembly has been the focus of considerable research in recent years because it can lead to superstructures with a complexity inaccessible by molecular self-assembly, and functionalities entirely different from or superior to those of the primary particles. Examples in molecular self-assembly suggests that anisotropic interactions could be useful in promoting particle self-assembly, with the exception of colloidal crystallization, which requires particles of uniform size and shape. Anisotropic particles prepared by surface modification of precursor particles are often rigid and submicron or micron sized, and thus relatively strong isotropic van der Waals interactions tend to resist self-assembly into regular superstructures. In addition, the relatively large contact area between particles needed for a sufficient binding enthalpy to stabilize a superstructure is difficult for rigid spherical particles. In contrast, flexible anisotropic polymeric particles dispersed in solvents have been shown to self-assemble into various superstructures. The flexibility of primary anisotropic particles enables them to fuse and stabilize into a superstructure. Some flexible and multi-component particles that are isotropic in common solvents can undergo deformation and sufficient material redistribution to anisotropically self-assemble into regular superstructures in selective solvents. The self-assembly is also driven by anisotropic interactions, which is induced during self-assembly rather than in the particles as synthesized. This review focuses on recent achievements in soft particle self-assembly and describes briefly the advancements in rigid particle self-assembly. The presentation is divided into discussion of self-assembly by the colloidal crystallization of isotropic rigid particles, anisotropic rigid particles, anisotropic soft particles and isotropic soft particles, in that order.

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## 1. Introduction

Self-assembly is a topic of intense scientific interest, as demonstrated by the inclusion of “How far can we push chemical self-assembly” in the Top 25 big questions discussed in the special 125th anniversary issue of *Science* magazine [1]. The past three decades have seen an explosive development in molecular self-assembly. Various strategies for molecular self-assembly have been developed for the construction of molecular aggregates with designed properties, geometries and dimensions. These promise to address or provide solutions to both theoretical and practical problems in many areas, such as drug delivery, catalysis, medical diagnostics and sensors [2–23]. In recent years, self-assembly using particles as building blocks, has drawn attention as a newly emerging scientific area. Self-assembly of particles controls the distribution of particles on the entire assembly by modulating inter-particle interactions on the nanoscale and focuses on constructing ordered and complex structures that are inaccessible to molecular self-assembly. Self-assembled superstructures can have functionalities that are different from or superior to those of the primary particles. For example, mono-dispersed silica and polymer latex were used as building blocks in colloidal crystallization to build photonic crystals with a tunable band gap by varying the stacking symmetry and inter-particle distance [24,25]. Gold nanoparticles decorated with linkers can be used for the detection of a variety of substances based on plasmon coupling by different nanoparticle aggregation mechanisms [26]. Silica spheres with a low polystyrene grafting density can self-assemble into string or sheet-like aggregates in a polystyrene matrix, which are superior mechanical enhancers for the matrix [27]. Multi-compartment nanostructures, fabricated from the self-assembly of polymeric nanoparticles with delicately designed inter-particle interactions, can serve as templates for decoration of functional species on specific compartments [4,28].

Apart from helping us design and prepare novel superstructures and functional materials, studying particle self-assembly can help us understand more about how life works because particle self-assembly is an essential

process carried out within organisms. Proteins self-assemble intra-molecularly into particles, and particle–particle or particle–biomolecule self-assembly leads to hierarchical structures such as the cell membrane [29], chromatin [30,31], and microtubules [32].

This review describes recent scientific efforts toward understanding the self-assembly of particles and is divided into four parts according to the types of building blocks used. We begin by briefly surveying colloidal crystallization of isotropic rigid particles such as mono-dispersed silica spheres, polymer latex, semiconductor nanoparticles and metal nanoparticles with designed surface functionality, and charged inorganic nanoparticles. Colloidal crystallization of silica or polymer latex on the order of hundreds of nanometers is usually driven by entropy, while colloidal crystallization of smaller nanoparticles (approximately 10 nm in size) is mainly driven by isotropic inter-particle attractions such as van der Waals interactions, DNA base pair interactions and electrostatic interactions. In the second part of our review, we briefly discuss the preparation and self-assembly of anisotropic rigid particles. Much effort has been devoted to the preparation of anisotropic rigid particles with designed features, and a variety of these particles have been reported in the literature. In most cases, anisotropic rigid particles are submicron or micron sized, and it is difficult for them to self-assemble into regular superstructures. Strong van der Waals interactions caused by the relatively large mass counteract the anisotropic interactions, and this rigidity makes anisotropic rigid particles difficult to stabilize into superstructures. For the third part of our review, we summarize the self-assembly of anisotropic soft particles that are usually polymeric or have polymeric components and can be dispersed in solvent. This solvent dispersibility makes anisotropic soft particles able to self-assemble in solution, where the isotropic inter-particle van der Waals attraction is overcome by solvation of the polymeric component; the solvated polymer chains are repulsed by one another due to their exclusive volume. Spherical, cylindrical, tubular and sheet-like superstructures can thus be obtained. During self-assembly, anisotropic soft particles are able to adjust their shape and anisotropy due to their flexibility, which should be necessary for the formation

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