Self-Assembly of Nanoparticles: A Snapshot

Soon after the discovery of methods for nanoparticle (NP) synthesis, scientists observed the ability of nanoparticles to self-assemble.[1−5] Since that time, the area has evolved into a field with considerable phenomenological breadth. Compared to classical colloidal particles, which produce superlattices following motifs of close-packed spheres, nanoscale particles brought about exceptional diversity of assembly patterns.[6] To some degree, such diversity is not surprising. The reduction of particle dimensions from the micrometer scale to the nanoscale leads to interparticle interactions that are more subtle and variable than those typical for classical colloid chemistry. In fact, nanoscale particles start resembling self-assembly components found in biology, such as globular proteins,[7] and replicate their assembly behavior.[8] We can also see the unity of self-assembly phenomena across scales[9] and start applying formal theories of self-organization phenomena.[10] The early technological applications of NP self-assembly processes taking advantage of their intrinsic scalability are also emerging.[11]

Hierarchical assembly remains one of the most appealing targets in nanoscience,[12] with the idea that we can use NPs and other building blocks in assemblies so as to tailor the properties of the material produced. Since we started publishing *ACS Nano*, we have explored these ideas, laying out the challenges and opportunities in this field.[12−14] In the United States and elsewhere around the globe, some of these challenges have been taken up as national and international challenges.[15] We are proud to continue to publish many of the leading advances in this field.

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This issue of *ACS Nano* contains a wonderful sampling of ideas driving current studies of NP assemblies. A step toward development of new concepts in this area is described in the interesting study of the effects of symmetry on self-assembly patterns that points out the importance of competition in different assembly pathways.[16] In order to accomplish advances in the fundamentals of self-organization, one also needs better understanding of NP anisotropy, which often defines which of many assembly patterns will dominate. Therefore, studies on the atomic-level design and metrology of Au clusters[17] and metal-tipped semiconductor nanorods[18] deserve particular attention.

Self-assembled systems made from plasmonic NPs display strong sensitivity to even slight changes in the geometry of the assembled structures. The variety of such systems continues expanding despite being among the earliest studied cases of self-assembly. Continuing expansion is fueled, in part, by the discoveries of new materials with plasmonic bands in the IR part of the spectrum, represented here by the study of the plasmonic effects for MoS_2 NPs.[19] The technological importance of plasmonic assemblies can be found in the study of Au/Ag nanocubes for solar cells.[20] The roles of similar systems for biosensing and imaging also constitute parts of the two Perspective articles published in this issue.[21,22]

The relevance of dynamic systems made from NPs and globular proteins becomes particularly clear in studies of surprising *in vivo* instability of Au−Ag nanoshells[23] and biological effects of MnO_2 NPs with albumin.[24] These studies provide a new point of view on NP behavior in biological environments. The analogy between the behavior of proteins and some NPs can be unmistakably identified by observing integration of NPs and their clusters in vesicle bilayers[25] and clearance of NPs by the reticular endothelial system.[26]

In the past decade, we have also seen rapid growth in the area of NP self-assembly for energy conversion and storage. As such, simple motifs of NP assemblies with cricoid proteins are shown to benefit energy harvesting.[27] The widespread interest to conductive hybrid materials incorporating NPs is impossible to overlook. It originates from the ability of

Published online April 22, 2014
10.1021/nn502057r

inorganic NPs to transport/store electrons and to self-organize into charge conduction pathways.[28,29] Recently, we have seen special attention given to self-assembly of graphene-based composites with earth-abundant NPs. Although there are no intricate three-dimensional assembly motifs observed for such systems yet, the ability of graphene and other two-dimensional materials to template NP formation provides a reliable and versatile method of controlling their organization. This issue features new studies in this field devoted to hybrid graphene−$CoSe_2$ catalysts for water oxidation[30] and Fe_2O_3−graphene superstructures for lithium batteries.[31] Along similar lines, charge trapping accompanying charge transport in NP solids is critical for their performance. For everyone interested in photovoltaic applications of NPs methods to improve performance, we recommend reading the new study on the trap-induced losses in NP−polymer materials.[32]

Last but not least, the research on self-assembling semiconductor NPs evolves toward their integration with well-developed micro- and mesoscale lithography tools.[33,34] This research is driven by the technological logic, and the success of such integration is likely to determine the viability of NP films in new platforms for electronics. The advances in this area can be found in the paper describing perfect ordering in sub-100 nm NP arrays molded by ultrathin alumina membranes.[35]

Disclosure: Views expressed in this editorial are those of the authors and not necessarily the views of the ACS.

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