University of Nebraska Lincoln | NEBRASKA CENTER FOR ENERGY SCIENCES RESEARCH

Cycle 11
January 2017

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Self-Assembled Meso-Architectures: Hierarchical Energy Materials from Nanoscale Components

Abstract. Nanoparticle self-assembly into mesoscale superstructures is a promising approach for constructing novel energy materials and systems from nanoscale components. Existing approaches for studying self-assembly, limited to assessing the final product ex-situ or tracking long-range order in reciprocal space, are insufficient for realizing advanced capabilities such as the development of adaptive materials or hierarchical bio-inspired mesoscale architectures. The fundamental understanding and control needed to address such challenges require novel approaches for probing self-assembly processes in real space in the native liquid environment. Here, research is proposed that will establish liquid-cell electron microscopy as an in-situ probe of the fundamental mechanisms, pathways, and forces that govern self-assembly. Liquid-cells with complex fluidic capabilities for controlled release and mixing of solutions will be conceived to perform systematic experiments over a wide parameter space and to actively influence the assembly process while observing the outcome in real time. Novel protocols will be developed for imaging the assembly of nanoparticle-based materials with single-particle resolution in solution. And the capability to prepare nanostructures with programmable interaction encoded in DNA will be established. These novel tools will be applied to investigate the assembly of ligand- and DNA-terminated nanocrystals, and the reconfiguration of nanoparticle superlattices in response to external stimuli. The expected outcomes from this project – a unique in-situ probe of self-assembly and an enhanced fundamental understanding of the processes underlying assembly and reconfiguration of nanoparticle superstructures – will pave the way for developing complex, dynamically tunable energy materials and mesoscale systems whose functionality approaches that found in biological energy conversion complexes. This long-term goal will be pursued within the framework of a center-scale (DOE-EFRC) research program that will be initiated on the basis of the novel capabilities, proofs of principle, and research team developed and organized under this project.