ASSEMBLY OF COLLOIDAL NANOCRYSTALS INTO OPEN NETWORKS

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Inorganic nanocrystals exhibit a wide variety of optical, electronic, chemical, and electrochemical functionality that is synthetically tunable based on their size and composition. Their properties and emerging methods for functionalizing their surfaces with specific chemical agents pose the exciting prospect to program the assembly of nanostructured materials whose properties depend intimately on both the characteristics of the building blocks and the mesoscale organization of these in the assembly. In this presentation, I describe novel strategies for assembling optically active nanocrystals into organized gel networks. In particular, theoretical frameworks predict open gel architectures when the extent of inter-particle bonding (i.e. valence) is constrained.[1] To achieve a chemically tunable valence, we functionalized semiconductor nanocrystals with highly charged chalcogenidometallates clusters that induce long range repulsive interactions.[2] The addition of controlled amounts of a cationic crosslinking agent determines the assembly of the nanocrystals into a low volume fraction gel. In another assembly strategy, short range attractive forces are induced between charge-stabilized nanocrystal colloids by the introduction of oligomeric polyethylene glycol (PEG). At low PEG concentrations, it can crosslink nanocrystals into a gel. At higher concentrations, PEG effectively passivates the nanocrystal surfaces, yet excess PEG can induce gel network assembly through the depletion attraction. The organization of the gel networks is characterized by small angle X-ray scattering, from which the fractal dimension that describes the gel topology is determined. We present an integrated approach leveraging theory, synthesis, characterization, and simulation to predict, realize, and analyze the formation of low volume fraction gels from colloidal nanocrystals with unusual optical properties in the visible and infrared spectral ranges.

References:
