

MAR16-2015-000456

Abstract for an Invited Paper  
for the MAR16 Meeting of  
the American Physical Society

**Polymer Grafted Nanoparticle Assemblies: From Optical to Mechanical Performance through Clusters, Monolayers and Monoliths<sup>1</sup>**

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Solution or melt-based fabrication of large area, matrix-free, ordered assemblies of polymer grafted nanoparticles (PGN) will enable additive manufacturing of novel membrane, electronic, and photonic elements. Due to the single component nature of these hybrids, aggregation and phase separation common in blended polymer nanocomposites are avoided. Architecturally, PGNs combine characteristics of colloids, brushes and high molecular weight polymers. Thus the processing-structure-property relationship of the entangled PGN assembly is unique from analogous condensed nano-structures, such as ligand stabilized nanoparticles, hard-sphere colloids, star macromolecules and linear chain – nanoparticle blends. Here in, we will discuss the intermediate character of PGNs with respect to deformability, physical aging, and rapid fabrication of stable, large-area, ordered PGN monolayers. For example, processing via flow coating follows that of classic colloids; however local structure and order within the PGN assembly is determined by the canopy architecture and substrate interactions. From this insight, large-area (cm<sup>2</sup>), highly-ordered, monolayer polystyrene-Au nanoparticle films that are resistant to de-wetting can be fabricated on substrates with high interface energy (80 mN/m) within seconds using flow-coating and a volatile solvent (THF). Overall these findings imply intriguing parallels between PGN assemblies and other mesoscale ordered polymeric systems including hard-soft block copolymers and semi-crystalline polymers. With the appropriate corona architecture, PGNs afford opportunities to design high inorganic fraction hybrids that retain processibility and enable the creation of films and fibers for next generation optoelectronic applications.

<sup>1</sup>Acknowledgement: Justin Che, Christopher A. Grabowski, Yang Jiao, Ming-Siao Hsiao, Kyoungweon Park, Lawrence Drummy