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Title: Understanding the role of multivalent counterions in the interfacial self-assembly of tripeptides when introduced to differing surface chemistries

Abstract: The self-assembly of peptides and peptide-conjugates has numerous applications ranging from tissue scaffolds to biocatalysis, but the role of multivalent counterions in the morphology of the hierarchical nanostructure remains unclear. This work seeks to understand how multivalent counterions interact with a simple model tripeptide to understand the role of counterions in self-assembly in the bulk versus at a surface. Our work focuses on short tri-amino acid peptides. These peptides offer a unique system to examine the impact on how each amino acid interacts leading to a better understanding of how rational sequence design can manifest in the formation of peptide-based materials. In this work, the tripeptide HYF (histidine-tyrosine-phenylalanine) is used to examine the self-assembly process at the interface of amine terminated and carboxylate terminated polystyrene nanoparticles while in the presence of multivalent cationic counterions. Self-assemblies were quantified by dynamic light scattering, zeta potential, and Anomalous Small Angle X-ray Scattering (ASAXS). The self-assemblies show an increased hydrodynamic radius while maintaining the same zeta potential as the bare nanoparticles, which suggests that adsorption of HYF onto the nanoparticles surface is a result of layered peptide:ion complexes at the surface. The incorporation of cationic counterions can also be seen by resonant trace of the ASAXS signal. ASAXS results define the morphology of the self-assembled tripeptides, showing a multilayered profile of the HYF with intercalated cation layers. By altering the surface chemistry of the we hope to illuminate the relationship between peptide/ion self-assembly in the bulk and epitaxial and growth self-assembly from a surface.