

Guo, C., Arnon, Z.A., Qi, R., Zhang, Q., Adler-Abramovich, L., Gazit, E., Wei, G. Expanding the Nanoarchitectural Diversity Through Aromatic Di- and Tri-Peptide Coassembly: Nanostructures and Molecular Mechanisms. *ACS nano*; 2016: 10, 8316-8324.

**Abstract:** Molecular self-assembly is pivotal for the formation of ordered nanostructures, yet the structural diversity obtained by the use of a single type of building block is limited. Multicomponent coassembly, utilized to expand the architectural space, is principally based on empirical observations rather than rational design. Here we report large-scale molecular dynamics simulations of the coassembly of diphenylalanine (FF) and triphenylalanine (FFF) peptides at various mass ratios. Our simulations show that FF and FFF can co-organize into both canonical and noncanonical assemblies. Strikingly, toroid nanostructures, which were rarely observed for the extensively studied FF or FFF, are often seen in the FF-FFF coassembly simulations and later corroborated by scanning electron microscopy. Our simulations demonstrate a wide ratio-dependent variation of nanostructure morphologies including hollow and solid assemblies, much richer than those formed by each individual moiety. The hollow-solid structural transformation displays a discontinuous transition feature, and the toroids appear to be an obligatory intermediate for the structural transition. Interaction analysis reveals that the hollow-solid structural transition is mostly dominated by FF-FFF interactions, while the nanotoroid formation is determined by the competition between FF-water and FFF-water interactions. This study provides both structural and mechanistic insights into the coassembly of FF and FFF peptides, thus offering a molecular basis for the rational design of bionanomaterials utilizing peptide coassembly.