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| Title: | Regulating Peptide Self-assembly toward precision & Compartmentalization |
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| Abstract: | Supramolecular assemblies are essential for specific biological functions and mandate precise control over the mesoscopic scale for higher functional efficiency. Such well-defined bio mimetic self-organization can be accessed through kinetically controlled non-equilibrium transformations. Recently, spatio-temporal control for the living supramolecular polymerization has rendered a paradigm shift towards designing complex multi-component supramolecular active materials; however directing the active monomer towards predictive kinetically trapped materials still remains a considerable challenge as that necessitates circumventing spontaneous nucleation of the monomers during the self-assembly process. We demonstrate dual strategies (chemical and photo) to sequester the active peptide self- assembling motifs in off-pathway dormant states that upon judicious activation, engage in controlled seeded supramolecular polymerization in aqueous milieu. The non-equilibrium responsive peptide self-assembly coupled with dormant metastable states allow access to interesting repertoire of structural diversities and attenuated biological functions with controllable supramolecular peptide nanostructures. Further, we explored the redox responsive peptide amphiphiles where tweaking the size of hydrophobic carbon chain between redox responsive ferrocene and self-assembling peptide fragment can leads to different self-assembled nanostructures in water. Changing the polarity of ferrocene moiety on treating with redox agent provides control over fine tuning of the assembly-disassembly behaviour. Finally, such control was efficiently used towards designing mixed block co-fibers of oxidation responsive periphery with rather intact non redox responsive cores as mediated by seeded supramolecular polymerization. Supramolecular self-assembly is of vital importance to realize excellent ability of nature to make complex designs with self-organization and self-sorting of simple molecules. In this context, we studied the fate of peptide self-assembly towards self-sorting or co-assembly with tiny structural mutations. We envisage the fidelity and stereo-selectivity of the chiral self-sorting by FRET with pyrene-hydroxy coumarin donor-acceptor pair tethered to the peptide sequences. Seed promoted elongation of the homochiral peptide amphiphiles through AFM image analyses and Thioflavin- T (ThT) binding validated the chiral recognition of the L/D peptide nanofibers. Moreover, direct visualization of the chirality-driven self-sorted nanofibers are reported through super resolution microscopy that exhibits enantioselective enzymatic degradation for L-peptide fibers. In the final chapter, the fidelity of fluorescent dye coupled peptide amphiphiles having varied carbon spacer between bisurea bonds was investigated towards co-assembly, self-sorting and co-block formation. Matching spacer resulted into randomly co-assembled fibers, whereas non-matching spacer length leads to self-segregate that shows no region of overlapping as monitored through super resolution microscopy. Finally, we have directly visualized the block formation through seeded supramolecular polymerization with same spacer size and distinct fluorescent dyes. |
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