



The Cross-scale Potential of Self-Assembling Peptides – Programming Molecular Information for Assembly to Nanofibril Architectures and Soft Matter Functions

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Molecular information gives rise to functional supramolecular assemblies across length scales in nature. Inspired by this programmability of cross-scale structural complexity of biological soft materials, self-assembling peptides (SAPs) were molecularly programmed in their supramolecular organisation via establishment of discrete interaction motifs. This anchors the hierarchically orchestrated assembly into peptide nanofibrils (PNFs) with complex precision architectures such as multichannel nanofibrils in the molecular identity and can even transcend from the molecular level into the macroscopic regime, where SAPs can introduce reversible cross-links as PNF gelators to create soft materials and extracellular-matrix-like scaffolds. This strategy offers a promising approach to dynamic cell environments similar to those found in nature and creates functional soft materials that overcome limitations that current systems frequently exhibit the material scale, such as non-balanced level of physical crosslinking yielding materials with insufficient mechanical profiles or static crosslinking, that is opposed to native dynamic cell-matrix interactions, enabling cross-scale innovation in biofabrication and -medicine. Furthermore, the combination of biopolymer-derived backbones with such SAP gelators combines their programmability of supramolecular dynamic interactions and multifaceted structure formation with mechanical stability offered by the backbone to enable new functions in rationally tailorable and broadly tunable 3D soft materials as sustainable alternatives to classical, synthetic polymer-based hydrogels. In the presented systems, the bio-inspired interactions give rise to excellent mechanical and biochemical properties, such as shear-thinning and autonomous recovery of supramolecular crosslinks as well as cytocompatibility with multiple cell-types. As important prerequisite, these features enable the development of biopolymer conjugates, so-called biohybrids, as bioinks for next-generation 3D bioprinting for different printing approaches. The necessity for functional 3D soft materials that combine scalable molecular precision, reproducibility, biocompatibility, and advanced and tailorable mechanical properties has grown steadily. Overall, the presented SAPs and PNF systems give rise to functional soft materials and act as platforms opening various application scenarios based on the cross-scale potential of self-assembling peptides as precision molecules.

