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Title: Development of Highly tunable Tunable bioactive peptide hydrogel scaffolds for applications in energy and healthcare

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Abstract:

Peptide self-assembly is a powerful bottom-up strategy for the synthesis of nanomaterials with complex, hierarchical architectures. Peptide nanomaterials have found interesting applications as advanced materials in biomedicine, tissue engineering, renewable energy, environmental science, nanotechnology and material science, etc. Comprehensive studies have revealed that the final function and application of the nanomaterial is dictated by its structural features at nanoscopic scale. Thus, the control over the self-assembled structure formation is the critical parameter in fabricating the desired functional material, which demands fundamental understanding of the self-assembly process. In this work, we exploited the self-assembled peptide hydrogels as an efficient synthetic extracellular matrix, providing the structural support as well as bio/physicochemical cues to control the stem cell behavior. To this direction, we initially explored the non-conventional approach of using different self-assembly pathways to create diverse nanostructures instead of changing the molecular architecture of the building blocks. Our study emphasizes on the role of self-assembly pathway in accessing diverse hydrogels from identical gelator concentration. Interestingly, we were able to modulate the biological response of the cells towards nanomaterial by tuning the nano structural morphology. Such observation led us to envisage that the differential self-assembly pathways would be a superior strategy that have the potential to develop a plethora of functional materials, and could eliminate the synthetic challenges towards developing new materials. Further, in the direction of controlling peptide self-assembly we even explored the effect of modulation of pH as a potential self-assembly pathway in governing the physicochemical properties of the final gel phase material. Here, we used a classical non-gelator with the ionic complementary sequence FEFK, which was rationally conjugated to an aromatic group naphthoxyacetic acid (Nap) at the Nterminal end to tune its gelation behavior. Depending on the overall charge of the exposed surface of the peptide amphiphiles as a function of pH, we were able to access diverse self-assembled nanostructures within a single gelator domain. The presence of oppositely charged amino acids in the peptide amphiphile resulted in pH-responsive behavior, leading to the formation of hydrogels over a wide pH range (2.0-12.0). To explore minimalistic biomimetic approach, we designed self-assembling N-cadherin mimetic short peptide which can perform the function of whole protein. The bioactive peptide hydrogel scaffold able to mimic biological properties of native protein like cell adhesive, proliferative and migration property. The next step was intended to develop bioactive peptide cellulose hybrid hydrogel by simply mixing the nanofibrous cellulose with the N-cadherin mimetic peptide. This hybrid hydrogel combines the unique characteristics of each component and overcome their individual limitations, such as biocompatibility and low stiffness, thus creating a suitable scaffold for regulating the cellular behaviour. Furthermore, we have developed peptide hydrogel based efficient charge transfer material, which can have potential application in artificial energy harvesting systems. We were able to tune the charge transfer efficiency by applying the biocatalytic control over the self-assembly process of the donor conjugated peptide molecule. The present work highlights the development of tuneable functional materials, which can provide essential biochemical and biophysical cues for controlling cellular behaviour. The non- conventional approach of modulating the self-assembly pathway to generate diverse biomaterial hold great potential to create advanced nanomaterials with desired function

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