Title:	DESIGNING BIOMOLECULAR GELS FOR APPLICATIONS IN ENERGY AND HEALTHCARE
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Abstract:	In recent years, supramolecular nanoarchitectures fabricated from bioinspired materials are gaining extensive attention due to their unique properties and wide applications in different fields of energy and healthcare. These bio-inspired smart materials generated by classical biomolecular self-assembly are primarily governed by different non-covalent interactions among specific building blocks. Such unique strategy of biomolecular assembly can create advanced functional materials with emergent superior properties. To this direction, natural carbohydrate-based polymers, like cellulose, glycosaminoglycans, etc. as well as short peptides or proteins are emerging as unique classes of bioinspired materials that can form advanced biomolecular matrices and show their potential in different domains: energy as well as healthcare. Over recent years, cellulose has been recognized as one of the bioinspired, renewable, and sustainable polymers which is emerging as one of the most important next-generation materials in the energy domain. To this end, we have developed a facile strategy of synthesizing cellulose nano-fibers from biomass-derived wood pulp by TEMPO-oxidation and incorporated iron oxide nanoparticles to synthesize nanohybrids for thermal insulation applications. Interestingly, in these nanohybrids, functional attributes like mechanical strength and flammability were improved to a greater extent, which overcomes the limitations of the commercially available thermal insulators in terms of their stability and durability. Most importantly, the nanohybrids demonstrated very low thermal conductivity as low as 0.024 W m-1 K-1, indicating better insulating potential of these nanohybrids as compared to other conventional insulating materials. Additionally, we have also functionalized cellulose nano-fiber by fly-ash to make cellulose nanohybrids. This strategy will offer a dual advantage in transforming waste materials into wealth, which may be coupled with the potential to serve in the energy sector. Interestingly, the inclusion of fly-ash resulted in a significant decrease in the thermal conductivity of the nanohybrid to 0.034 W m-1 k-1. Thus, we envisage that these biomass-derived biomolecular composites can be used as thermal insulators to make energy-efficient smart buildings in the future. Furthermore, it is interesting to note that these cellulose nano-fibers are also evolving as an efficient template for their enormous potential in biomedicine. On the other hand, another bioinspired material i.e., peptides are also emerging as interesting candidates for developing bioinspired scaffolds that can be applied in diverse domains of healthcare. Specifically, short peptides inspired from bioactive proteins are being recognized as crucial viibuilding blocks that have gained much attention owing to their inherent bioactivity as well as unique physical, chemical, and biocompatible attributes. In principle, the formulation of peptide-based biomolecular gels is primarily driven by different non-covalent interactions that are quite sensitive to external stimuli such as pH, temperature, ions, etc. We can create diverse nanostructures within a single gelator system by controlling the external stimuli which can be effective in controlling cellular behaviour. In this direction, we have created Cbz-FF-OH dipeptide-based diverse nanostructures in a single gelator system by varying the thermal history from 40-90°C that could exhibit distinct differences in structure at the nanoscale as well as trigger differential cellular response in a single gelator domain. Furthermore, a step forward to mimic the diverse biomolecular entities present in the native extracellular matrix (ECM), we have demonstrated the non-covalent interactions of different carbohydrate-based biopolymers with short peptides as an emergent approach to create advanced biomolecular composite scaffolds. To this end, we have designed a biologically relevant positively charged Cardin-motif peptide that has the ability to bind negatively charged glycosaminoglycans which are present in the ECM. It has been observed that the designed peptide failed to form stable hydrogel at physiological pH owing to the presence of positively charged amino acids. In this direction, we have demonstrated Cardin-motif peptide and heparin-based biomolecular scaffold by employing simple non- covalent interactions between peptide and heparin at the molecular level. This biomolecular scaffold displayed tuneable nanofibrous morphology and superior biocompatibility towards neural cells as compared to that of the peptide scaffold. Interestingly, these composite hydrogels demonstrated superior gel stability, cellular viability as well as cellular proliferation in 3D culture conditions as compared to peptides. In a similar direction, we have also explored another multicomponent hybrid scaffold by mixing this bio-inspired peptide and plant-based biopolymer i.e., cellulose. Interestingly, this composite scaffold showed significant enhancement in cellular growth and proliferation of the fibroblast cells as compared to the only peptide. Furthermore, we have employed another non-conventional approach to overcome the limitations of positively charged Cardin-motif peptide that failed to self-assemble at physiological pH. We used an elegant strategy by employing different anions to the positively charged Cardin-motif peptide to mask the overall surface charge. We have demonstrated the different self- assembly behaviour of the Cardin-motif peptide in the presence of different anions, HPO42- viii, Cl-, and I-. Interestingly, these anions demonstrated remarkable effects in modulating the nanostructure formation and the mechanical stiffness of Cardin-motif peptide hydrogels owing to their differential interaction with water molecules according to the Hofmeister series. More importantly, these ion-coordinated stable hydrogels showed diverse cellular behaviour towards different cell lines such as fibroblast and neuronal cells. Our overall findings emphasize that the co-assembly approach to fabricate multicomponent hybrid systems can be envisaged as a beneficial approach towards overcoming challenges associated with the single component systems and can be more useful for applications in the energy and healthcare sectors.
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