Disordered proteins: A new avenue towards hierarchical functional materials

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There is growing evidence that intrinsically disordered proteins (IDPs) play a fundamental role in mineralization. IDPs contribute in intermolecular interactions at the protein–mineral interface. Here we report a protein-mediated mineralization process that takes advantage of disorder-order interplay using elastin-like recombinamers (ELRs) to program organic-inorganic interactions into hierarchically-ordered mineralized structures. During crosslinking process, ELRs self-assemble into a dense network of nanofibers and homogenously distributed three-dimensional (3D) ELR spherulites. Upon incubation in a solution supersaturated with respect to apatite, the materials comprise elongated apatite nanocrystals that are aligned and organized into microscopic prisms, which grow together into spherulite-like structures hundreds of microns in diameter. Given, the vast clinical need and potential impact of engineering more efficient materials to replace lost/diseased enamel, we conducted in vitro proof-of-concept studies to investigate the potential use of the hierarchical mineralized structures for dentin hypersensitivity as a mineralizing bandage to occlude exposed dentinal tubules. We confirmed that the hierarchically mineralized membranes grew, adhered, and conformed to the surface of the etched dental tissues. Integration between the hierarchical structures and the dental tissues was observed at the dentin-membrane interface, where the nanocrystals infiltrated and blocked dentinal tubules. The mineralized structures exhibited comparable acid resistance to dental enamel. Our approach takes advantage of the disordered nature of ELR molecules to trigger a supramolecularly organized organic framework capable of controllably templating the growth of apatite crystals at multiple length scales. This mechanism goes beyond biomimicry and opens up the possibility to not only modulate mineralization but also to explore ways of utilizing disorder-order interplay for the generation of functional materials. The study represents a potential strategy for complex materials design that may open opportunities for hard tissue repair and provide insights into the role of molecular disorder in human physiology and pathology.

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