STRUCTURE-PROPERTIES RELATIONSHIPS OF MULTICOMPONENT POLYSACCHARIDE-PEPTIDE HYDROGELS

Ronit Bitton, Department of Chemical Engineering and The Ilse Katz Institute for Nanoscale Science &Technology, Ben-Gurion University of the Negev, Israel
rbitton@bgu.ac.il

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Over the past two decades, the potential of hydrogels as scaffolds for tissue regeneration has been widely explored due to their similarity to native extra cellular matrices (ECMs) and the ability to easily adjust both their physicochemical properties as well as their mechanical properties to meet the demands for tissue scaffolds and cell encapsulation. To better emulate the functionality of the natural ECM, current effort in the engineering of synthetic extracellular matrixes has focused on installing molecular features (proteins and bio-interactive polymers) within insoluble scaffolds, either by self-assembly or through covalent modifications of polymer or biopolymer networks.

Polysaccharides (e.g hyaluronic acid, alginate, and chitosan) – being non-toxic, hemocompatible, and relatively cheap– possess many of the favorable properties required for biomaterials. Combining polysaccharides and peptides for creating hydrogels for tissue engineering is of particular interest, due to the complementary properties of both molecules: Bio-functionality of the peptides on one hand and similarity to the natural ECM of the polysaccharides on the other. Apart from their direct role in cell interaction, peptide sequences may affect the hierarchical structural organization and mechanical properties of the resulting hydrogel, thus indirectly affecting the cellular response. The overall aim of our study is to develop a fundamental understanding of the structure-mechanical properties relations of multicomponent polysaccharide hydrogels used in tissue engineering applications

Here we present a systematic investigation of the effect of RGD- containing peptides on the hierarchical structure of polysaccharide-peptide hybrids (solutions and gels). Polysaccharide type, ligand incorporation method (covalently attached or self-assembled) as well as peptide nanostructure and amount were tested using advanced tools including small angle X-ray scattering (SAXS), electron microscopy and rheology.

Our results show that the fraction of the covalently bounded peptide, determines the behavior of a polysaccharide-peptide conjugates in solution, regardless of the specific nature of the polysaccharide. And that the peptides' ability to self-assemble in aqueous solution affects the spatial organization of the polysaccharide and the mechanical properties of the polysaccharide /peptide hybrid hydrogel, both when the peptide is covalently attached to the polysaccharide backbone and when peptide and polymer solutions are simply mixed together. These results indicate the importance of possible intermolecular interactions between the peptide and the polymer in determining the hydrogel's properties.

Our findings suggest that elucidating key factors involved in the structure-property relationships of these systems will improve our ability to design and prepare tailor-made scaffolds for a variety of applications.