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Biohybrid Polymer Hydrogels for Regenerative Therapies

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Abstract - Cell-instructive characteristics of extracellular matrices (ECM) resulting from a subtle balance of biomolecular and biophysical signals must be recapitulated in engineered biomaterials to facilitate regenerative therapies. However, hardly any currently available material allows for the independent tuning of the involved molecular and physical cues due to the inherent correlation between biopolymer concentration and material properties. Addressing the resulting challenge, we introduce a rational design strategy for ECM-inspired hydrogels based on multi-armed poly(ethylene glycol) and heparin, adapting a mean field approach to identify conditions at which the balance of elastic, electrostatic and excluded volume forces results in constant heparin concentrations within swollen polymer networks with gradually varied physical properties. This theoretically predicted decoupling of biochemical and mechanical gel properties was confirmed experimentally and utilized for heparin-based biofunctionalization schemes to afford cell adhesiveness and morphogen presentation. Using this approach for the modulation of distinct material characteristics, different particular combinations of matrix conditions were identified to effectively stimulate the pro-angiogenic state of human umbilical cord vein endothelial cells and the differentiation of human mesenchymal stem cells. Our study demonstrates the power of joint theoretical and experimental efforts in creating bioactive materials with specifically and independently controllable characteristics.