Star-PEG-Heparin-Polyelectrolyte-Hydrogels - Theory, Experiment, and Simulation

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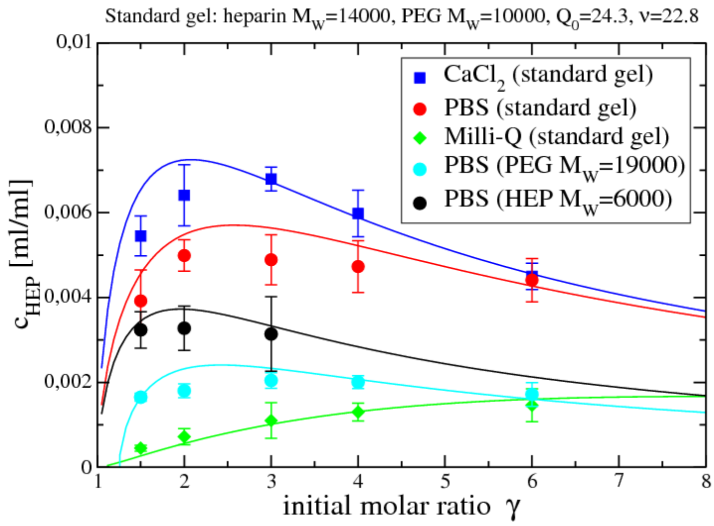

Fig. 1: Concentration of heparin in equilibrium swollen state (in ml/ml) in the experiment (filled symbols) compared with the theoretical model (lines). The blue, red, and green lines are the best fits of two model parameters covering the dominating effects. Hence, predictions are made for different building blocks (black and cyan lines) and confirmed by the experiment (black and cyan circles) showing a nearly constant heparin concentration under physiological conditions (PBS).

We are studying a new class of biohybrid hydrogels made of heparin, a rod-like highly charged glycosaminoglycan as a highly functionalized cross-linker, and non-charged, elastic 4-star-shaped-polyethylene glycol [1] by using experiment, theory, and simulation. We focus on the network structure by using the bond-fluctuation simulation method and determine the structural defects, the topological arrangement, and percolation threshold of the gels by varying the initial molar ratio, the functionalized groups, and the concentration of the reaction mixture. Also, we developed a mean-field type model to understand the combined effects of counter-ions and a good solvent on the swelling properties of the gels [2]. It has been shown that this interplay leads to nearly constant heparin concentration in the swollen gel under physiological conditions (see Fig. 1), while large variations of the degree of swelling and the storage modulus are accessible. Furthermore, we are able to predict the degree of swelling and heparin level for different molar masses of the building blocks allowing a rational design of the hydrogels [3]. This gels allow a constant release of heparin-binding signaling molecules while independently controlling the mechanical properties to optimize matrices with both mechanical and biomolecular features for cell replacement-based therapies.

[1] U. Freudenberg; et al. A star-PEG-heparin hydrogel platform to aid cell replacement therapies for neurodegenerative diseases. Biomaterials, 30: 5049-5060, (2009).
[2] Sommer, J.-U., Dockhorn, R. et al., Swelling Equilibrium of a Binary Polymer Gel, Macromolecules 44, 981-986, (2011)
[3] U. Freudenberg, J.-U. Sommer et al. Using Mean Field Theory to Guide Biofunctional Materials Design, Advanced Functional Materials 22, 1391-1398, (2012)