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Tuning nanoscale viscoelasticity of polyelectrolyte complexes with multiple types of cross-links TIANZHU MA, BIAO HAN, Drexel University, School of Biomedical Eng, Sci and Health Systems, DAEYEON LEE, University of Pennsylvania, School of Engineering and Applied Science, LIN HAN, Drexel University, School of Biomedical Engineering, Science, and Health Systems — Mechanical properties of hydrogels are manifestation of cross-link type and density, fixed charges and water-polymer interactions. In this study, we revealed how different types of cross-links regulate the nanoscale viscoelasticity of polyelectrolyte networks. Ionically cross-linked PAH/PAA layer-by-layer complexes were modified to include covalent cross-links using EDC. AFM-nanoindentation and force relaxation were performed at various ionic strength (0.01-1M) and pH (1.5-5.5). As-assembled networks, held only by ionic cross-links, underwent >95% relaxation, dominated by cross-link breaking and re-formation. Addition of covalent cross-links increased the instantaneous modulus by 1.6-fold and attenuated relaxation to $\approx 80\%$ of net neutral states ($\text{pH} \geq 3.5$), as covalent cross-links provide additional elastic components. The network remained stabilized when all ionic cross-links were dissociated at $\text{pH} \leq 1.5$, whereby further attenuation to 31% in relaxation could be due to viscoelastic polymer conformational changes and fluid flow-induced poroelasticity. Taken together, this study demonstrates the potential of using multiple cross-linking types to tune the viscoelastic mechanisms in polyelectrolyte complexes.

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