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Viscoelastic Nanomechanics of Ionically Cross-linked Polyelectrolyte Networks BIAO HAN, Drexel University, DAEYEON LEE, University of Pennsylvania, LIN HAN, Drexel University — Understanding the mechanics of ionic polyelectrolyte networks is critical for applications where nm-to-um mechanics is the key to success. This study aims to reveal the roles of ionic cross-links and fixed charges in the viscoelasticity of layer-by-layer poly(allylamine hydrochloride)/poly(acrylic acid) microfilms, PAH/PAA, a complex held by pH-sensitive amine-carboxyl links. AFM-nanoindentation and force relaxation (tip R=12.5um) was performed at ionic strength(IS)=0.01-1.0M, pH=5.5-2.0 (pKa of PAA=2.3). When pH changes from 5.5 to 2.0, the films swell for 4x from densely linked, net neutral state to loosely linked, positively charged one. A >100x reduction in indentation modulus was observed at all IS, suggesting the dominance of decrease in cross-link density. In most states, more than 90% force relaxation was observed, where cross-link breaking/reformation likely dominates viscoelasticity. However, at pH=2.5 and IS=0.01M, when electrical double layer repulsion is important (Debye length=3nm), relaxation was about 60%, highlighting the contribution of fixed charges. In summary, this study revealed unique viscoelastic behaviors of PAH/PAA due to the pH- and IS-dependent cross-link and charge densities.

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