Title: Electrochemically-Controlled Mechanical Properties of a Polymer Nanocomposite

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Stimuli-responsive polymer nanocomposites hold promise for applications in a number of areas including control of cellular or protein adhesion on surfaces, drug delivery, separations, and biomimetics, among others. Electrochemical stimuli are particularly advantageous as they can be applied rapidly, reversibly, locally (i.e., at an electrode instead of throughout the bulk), and remotely, while maintaining a mild operating environment. Here we present the layer-by-layer assembly of an electroactive polymer nanocomposite thin film containing cationic linear poly(ethyleneimine) (LPEI) and 68 vol% of anionic Prussian Blue (PB) nanoparticles. Incorporation of these PB nanoparticles allows for electrochemical control over film thickness and mechanical properties. Electrochemical reduction of the PB doubles the negative charge on the particles, inducing an influx of water and ions from solution to maintain electroneutrality in the film; swelling and increased elastic compliance of the film result. Upon cycling of the applied potential between -0.2 V and +0.6 V (versus Ag/AgCl), reversible film swelling on the order of 2-10% was measured with spectroscopic ellipsometry and electrochemical atomic force microscopy. Reversible changes in the elastic modulus of the hydrated composite film on the order of 50% (from 3.40 GPa to 1.75 GPa) were measured with *in situ* nanoindentation of these composites in the fully immersed state. These results present a new framework for electrically modulating the stiffness of a composite. Further, this swellable nanocomposite system opens up the possibility of investigating disruptable percolative networks in which interactions between nanoparticles are turned on and off with an electrochemical trigger, resulting in more dramatic mechanical changes. This work serves as a starting point for further studies on mechanomutable coatings with potential future applications in micro- and nanoscale devices.