549d Continuous Polyelectrolyte Nanofilm Growth under an Applied Electric Potential

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Polymer nanofilms offer facile control over the physical, chemical, and biological character of a material surface. Adsorption from solution is a simple means of fabrication, but control over film thickness is limited due to rapid saturation. Thicker, multilayer films are possible, but only through many alternate exposures to solutions of complementary species. We show here that a modest electric potential difference applied between an adsorbing substrate and a counter-electrode, in the presence of a polyelectrolyte solution, can lead to nanofilm growth that is continuous, without apparent saturation; films of arbitrary and controllable thickness may thereby be realized in a single step. We observe this behavior for poly-L-lysine onto indium tin oxide, at substrate potentials exceeding a threshold of 0.56 ± 0.05 V (relative to a standard hydrogen electrode), using optical waveguide lightmode spectroscopy (OWLS). Film growth kinetics are initially very rapid and subsequently become linear with time. Linear growth may even be re-established following interruption by placement of a protein layer, suggesting possible applications in biosensing and bioelectronics. Films grown under an applied electric potential exhibit very modest desorption, but are somewhat unstable to removal of the potential. Chemically cross-linking films using the EDC/NHS method results in significantly improved stability. Atomic force microscopy images reveal films to be particulate, with the particle size slightly exceeding the polymer's hydrodynamic diameter. We find no evidence of electrochemical oxidation at the adsorbing surface, and suspect that secondary structure formation may play a role in the observed behavior.