40g Nanometer-Scale Structure and Erosion Profiles of Erodible Multilayered Polyelectrolyte Films

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We have used atomic force microscopy (AFM) and scanning electron microscopy (SEM) coupled with ellipsometry to characterize the physical morphology and surface structures of 100 nm thick multilayered films fabricated from alternating layers of a hydrolytically degradable polyamine and either sodium poly(styrene sulfonate) (SPS) or plasmid DNA. Films fabricated from these polyanions are initially smooth, as characterized by AFM. However, we find striking differences in the topography, structures, and thickness profiles of these two materials as a function of full or partial erosion under physiologically relevant conditions (PBS buffer at 37 °C). For films fabricated from SPS, AFM data are consistent with an erosion process that occurs uniformly without the generation of holes or pits over large, micrometer-scale areas. By contrast, films fabricated from plasmid DNA undergo dramatic structural rearrangements that result in polymer/DNA particles ranging in size from 50 to 400 nm. Additional characterization of these particulate structures by SEM suggested that they are interpenetrated with or fused to underlying polyelectrolyte layers on the silicon surface, providing a potential mechanism to manipulate the adhesive forces with which these particles are bound to the surface. The reasons for the differences in the behavior of erodible films fabricated from these two different polyanions are not yet clear. However, in the context of gene delivery, the presentation of condensed DNA nanoparticles at these surfaces may be advantageous with respect to stimulating the internalization of DNA by cells. Thus, a quantitative understanding of the factors influencing this structural transformation is of interest in both fundamental and applied contexts. Structure/property relationships and an analysis of environmental parameters that lead to these transformations, and can be used to control them, will be discussed.