Supplementary Material

Figure S1. Schematic comparing the growth mechanisms of films displaying linear and exponential growth. Linear growth (top) follows the classic model of film development where dipping in each polyethylene (PE) results in monolayer formation, creating a bilayer at the end of each cycle. In this growth mechanism the thickness of the film increases by the same amount with each deposition step. Exponential growth (bottom) does not grow proportionally. One or more of the PE (red) is able to diffuse into the pre-existing film base during its incubation step. In the subsequent rinse step, some of the PE that had diffused into the film is lost to the rinse solution. Incubation in the PE of opposite charge (blue) results in typical bilayer formation (**a**), however once the surface charge is reversed, the PE within the film can diffuse back through the film to the surface (**b**) where it forms additional complexes (**c**). Hence more than one bilayer is formed per deposition step. The amount of PE that can diffuse into the film is proportional to the pre-existing film thickness and therefore the film grows exponentially thicker with each deposition step.

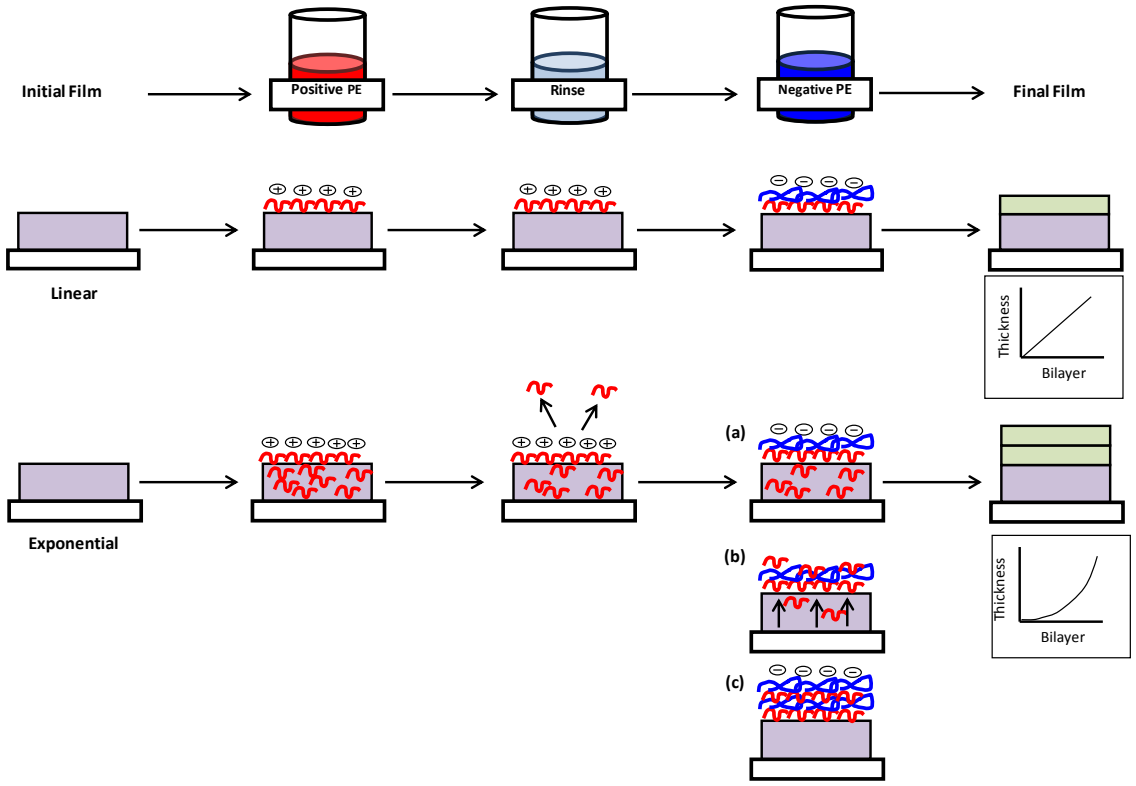

Figure S2. Schematic of the layer-by-layer assembly method for polyelectrolyte multilayers. A charged substrate (*i.e.*, quartz) is dipped in a polyelectrolyte of opposite charge (purple) where it adheres through electrostatic attraction, effectively reversing the surface charge. The film is rinsed with an appropriate solution to remove excess polyelectrolyte before dipping in the negatively charged polyelectrolyte (green). The negative polyelectrolyte adheres and the surface charge is again reversed. This process is repeated until a film of the desired number of multilayers is achieved.

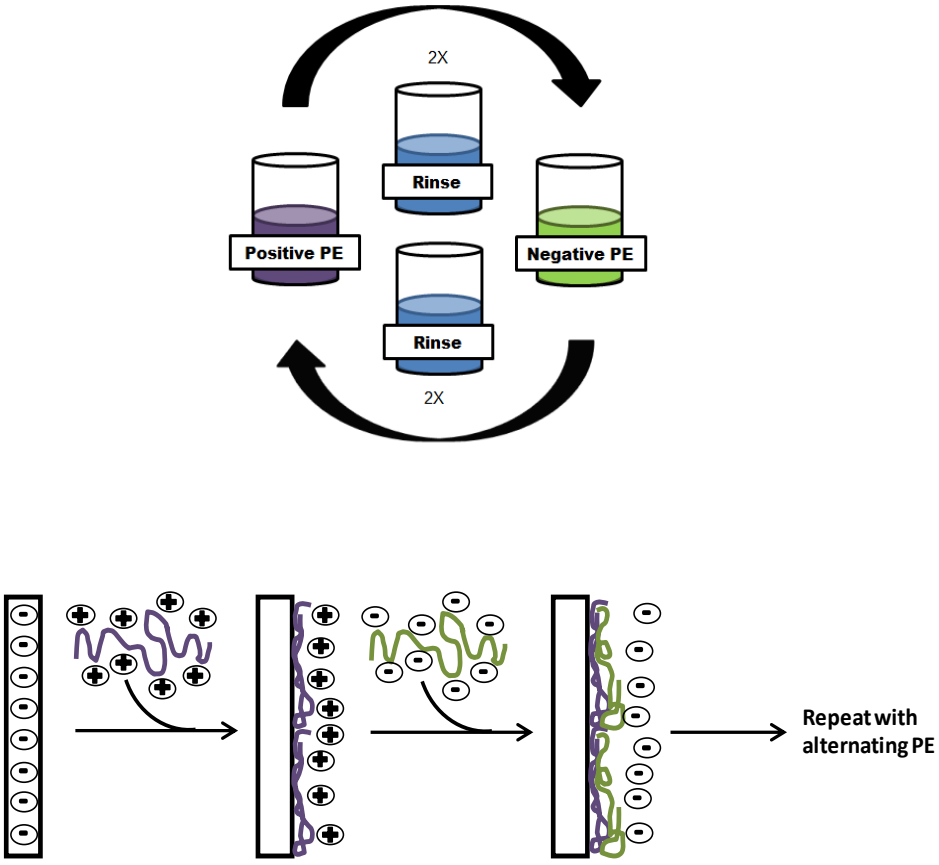

Figure S3. Representative AFM image showing surface topography of a 60 × 60 μm^2 square from a dry chitosan (CHI)/hyaluronan (HA)/sulforhodamine B aptamer (SA) film. (**a**) Height profile of selected area (white line) on a two dimensional (2D) image. Area highlighted in red represents the average peak height value showing the large variation in peak height. (**b**) Three dimensional (3D) representation showing overall film morphology.

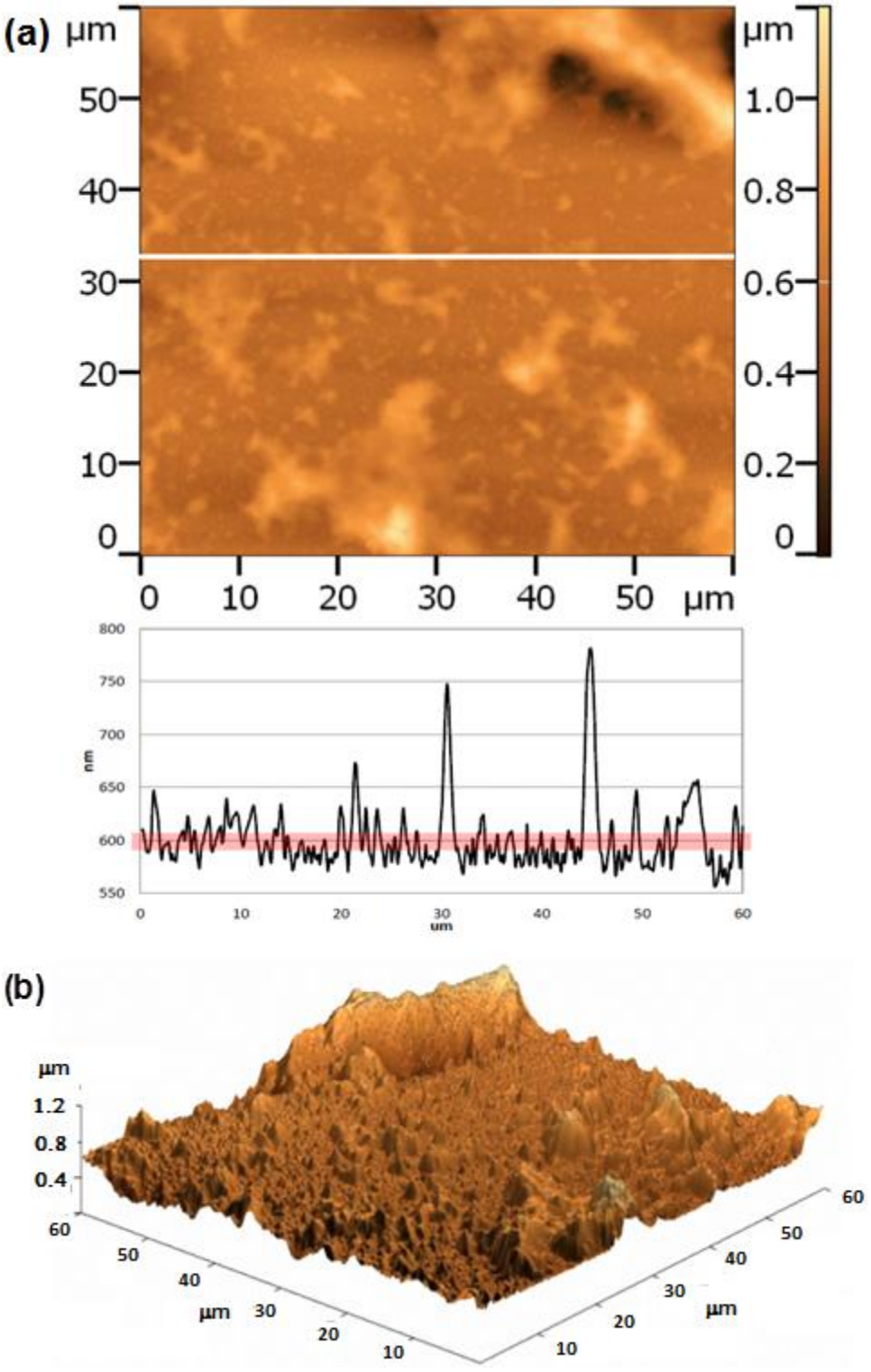

Table S1. Film morphology and roughness parameters determined for two 60 × 60 μm^2 AFM images of films containing the sulforhodamine B aptamer (SA) or RO control, with or without exposure to sulforhodamine B (SB) dye. Two trials (A and B) were taken from different areas on the same film. All values were calculated with NOVA software (Falköping, Sweden). Atomic Force Microscopy (AFM) images were taken in tapping mode.

Sample	Trial	Number points sampled	Average height [a] (nm)	Average roughness [b] (nm)	RMS roughness [c] (nm)	Average RMS roughness (nm)
SA (without SB)	A	262,144	601.9	82.3	119.4	157.1
	B	262,144	515.0	128.5	194.8	
SA + SB	A	262,144	640.7	157.7	193.4	198.1
	B	172,032	1,380.6	138.9	202.8	
RO (without SB)	A	262,144	460.4	113.6	143.7	185.9
	B	262,144	671.7	181.0	228.1	
RO + SB	A	262,144	508.7	96.0	134.4	160.1
	B	262,144	679.6	134.2	185.7	

[a] $\bar{z}(N,M) = \frac{1}{N}\sum_{x=1}^{N} z(x,y)$

[b] $R_a(N,M) = \frac{1}{N}\sum_{x=1}^{N}[z(x,y) - \bar{z}(N,M)]$

[c] $R_q(N,M) = \sqrt{\frac{1}{N}\sum_{x=1}^{N}[z(x,y) - \bar{z}(N,M)]^2}$