## Tunable stimulus-responsive friction mechanisms of polyelectrolyte films and tube forests

(Supplementary Information)

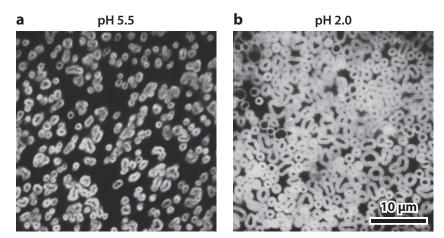
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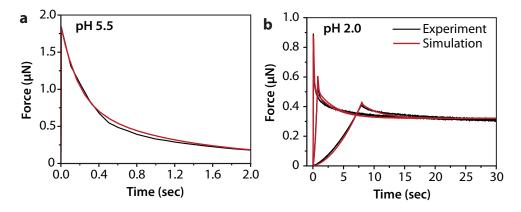
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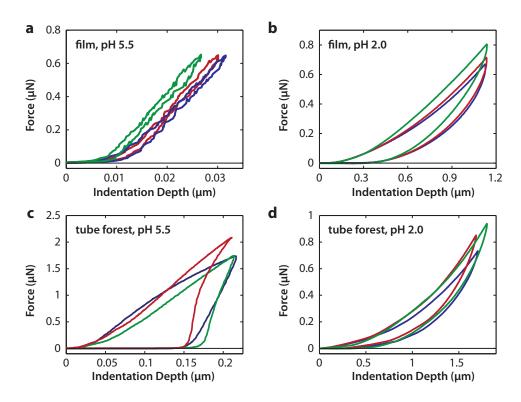
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**Fig. S1** Fluorescence images of the PAH/PAA tube forest at pH 5.5 and 2.0 (top view) (data adapted from ref. 24).



**Fig. S2** Time-dependent indentation behavior of PAH/PAA planar film using a hydroxyl-terminated selfassembled monolayer (OH-SAM) tip ( $R \approx 22.5 \,\mu$ m) in aqueous solutions, pH = (a) 5.5 (0.01 M NaCl) and (b) 2.0 (0.01 M HCl), and corresponding five-element spring-and-dashpot model fit. The force relaxation was measured after indentation at 1  $\mu$ m s<sup>-1</sup> rate at (a) pH 5.5, and at 10, 1 and 0.1  $\mu$ m s<sup>-1</sup> rates at (b) pH 2.0 (three individual relaxation curves and model fits). The corresponding model output of PAH/PAA moduli are (a)  $E_{\text{pH 5.5}} = 125(1-0.654(1-e^{-t/0.15})-0.293(1-e^{-t/1.0}))$  MPa, (b)  $E_{\text{pH 2.0}} = 83(1-0.5(1-e^{-t/0.1})-0.2(1-e^{-t/3.0}))$  kPa, where *t* is in seconds.



**Fig. S3** Typical indentation force-depth curves on PAH/PAA tube forest and planar film at 1  $\mu$ m s<sup>-1</sup> constant indentation-depth rate using a hydroxyl-terminated self-assembled monolayer (OH-SAM) tip ( $R \approx 22.5 \mu$ m) in 0.01 M NaCl (pH 5.5) and 0.01 M HCl (pH 2.0) aqueous solutions. Each of the three individual curves was taken at a different indentation location: (a) film, pH 5.5, (b) film, pH 2.0, (c) tube forest, pH 5.5, and (d) tube forest, pH 2.0. Negligible adhesion upon unloading was observed for all the cases.

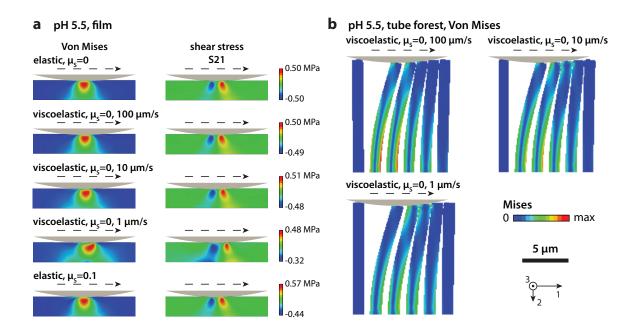
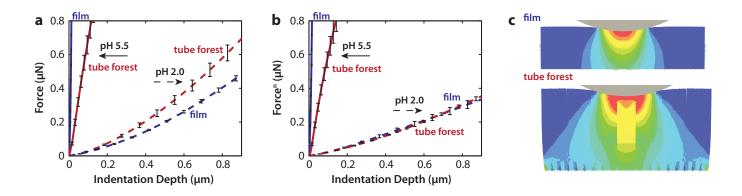


Fig. S4 (a) Comparison of von Mises and shear stress contours between elastic and viscoelastic continuous films at pH 5.5 and different lateral displacement rates. (b) Comparison of von Mises contours for the tube forest at pH 5.5 and different rates.  $\mu_s$  is the tip-sample surface friction coefficient.



**Fig. S5** Indentation force-depth curves on PAH/PAA tube forest and planar film at 1  $\mu$ m s<sup>-1</sup> constant indentation-depth rate (mean ± standard error of means,  $n \ge 10$  indentation locations), (a) original force versus depth curves, (b) substrate effect-corrected force (force<sup>n</sup>) versus depth curves, (c) finite element analysis predicted stress contour S22 of the film and tube forest at pH 2.0, data adapted from ref. 24.