

Supplementary information

pH-control of protein resistance of thin hydrogel gradient films

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Force measurements

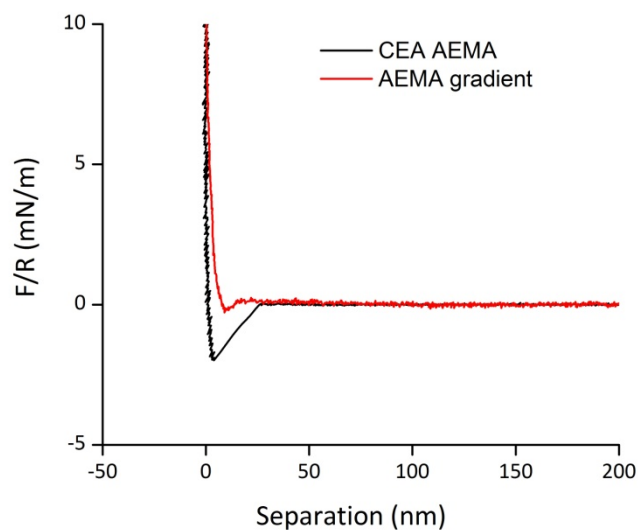


Figure S1. Interaction forces of a neutral ($C_{16}OH$ SAM-modified) probe interacting with a PAEMA layer (red) plotted together with the interaction measured at the AEMA-dominated end of a the P(CEA-AEMA) gradient (black) with a negative MUA probe. The black curve is the rightmost curve in Figure 4 at pH 6.4 where the attraction is greatest, and the total dry thickness of the polymer layer is the same in both samples. In this case the net force on both surfaces is attractive, but a quantitative comparison of the force curves is difficult, since the van der Waals and attractive electrostatic forces cannot be easily separated due to the fact that the probe “jumps” into contact with the surface when the gradient of the attractive force exceeds the stiffness of the cantilever, leaving very little data in the attractive region for fitting. The electrostatic interaction with the $C_{16}OH$ probe is weak due to the low charging of the $C_{16}OH$ surface. However, there is a short-ranged but significant steric interaction at about 10 nm separation in the interaction with the pure PAEMA layer, which is not present in the mixed layer. This indicates a compaction of the mixed film which is not present in the single component, as seen in Figure 5.

QCM-D data

Results are shown for PAEMA and PCEA in Figure S2 and Figure S3, respectively. PAEMA showed no mass uptake or change in dissipation at high pH (> 6), and was only marginally swollen at pH 5.3. At pH 4.3, there was a mass uptake and a positive shift in the dissipation representative of swelling. Interestingly, there response to pH changes in PAEMA is markedly slower than for PCEA. The reason for this is unclear, but might arise from a different ionisation pattern of the primary amine. The primary amine has high hydrogen bonding capabilities even in its deionised state, which might lower the driving force of the primary amine to be charged, but a complete explanation is, however, difficult to give. PCEA showed an increasing swelling upon raise in pH, with the largest shifts occurring between MQ and pH 7.0. This was also expected, as more carboxylic acids became ionised as the pH was increased. The data correlates well with the ellipsometry data, where the largest swelling response was seen at pH between 6.1 and 7.0, and only marginally increased swelling between pH 7.0 and 8.0.

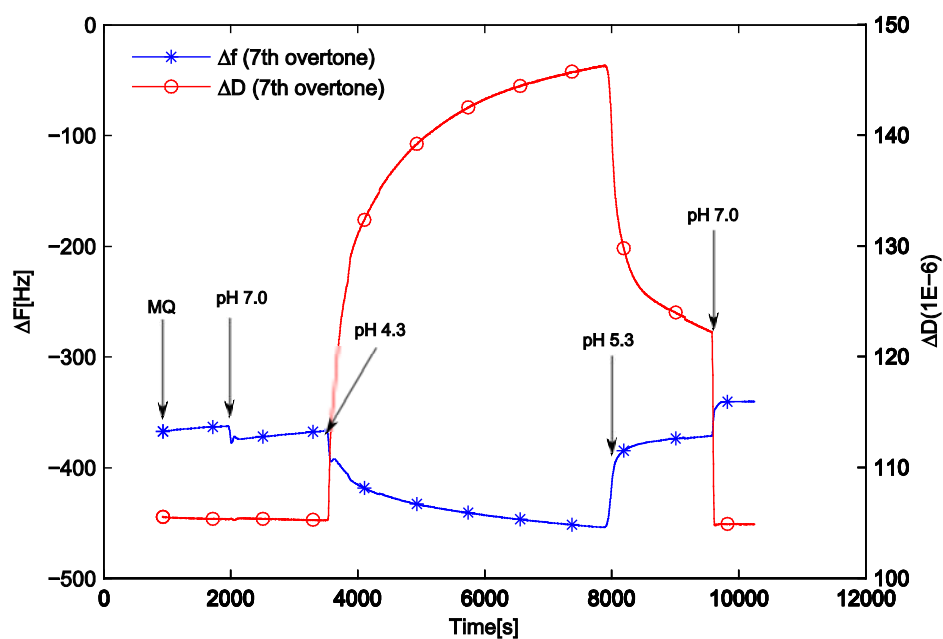


Figure S2. QCM-D data for PAEMA pH-dependent swelling.

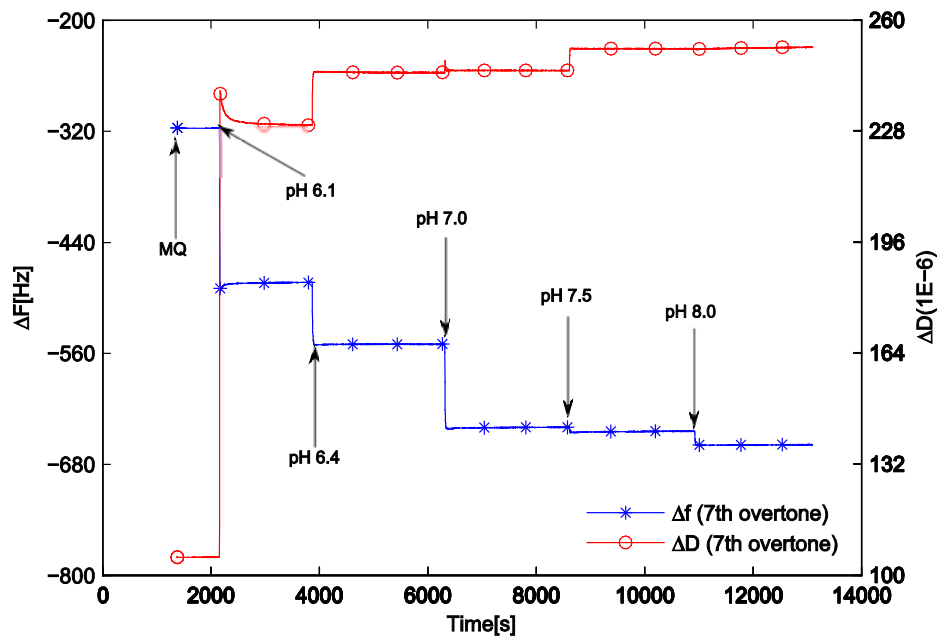


Figure S3. QCM-D data for PCEA pH-dependent swelling.