

Supporting information

Interaction Forces on Polyampholytic Hydrogel Gradient Surfaces

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Infrared spectra and peak assignments for MAA-DMAEMA and DMAEMA gradients.

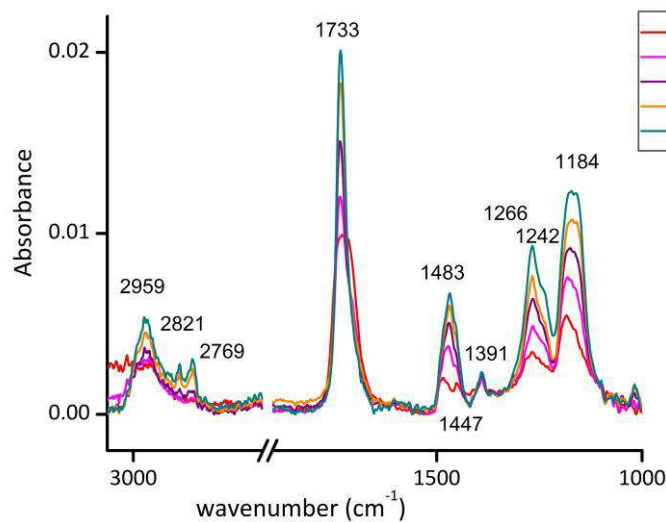


Figure S1. Infrared spectra along the P(MAA-DMAEMA) gradient. The positions A-I correspond to the positions indicated in Figure 3 in the main text. See Table S1 for peak assignments.

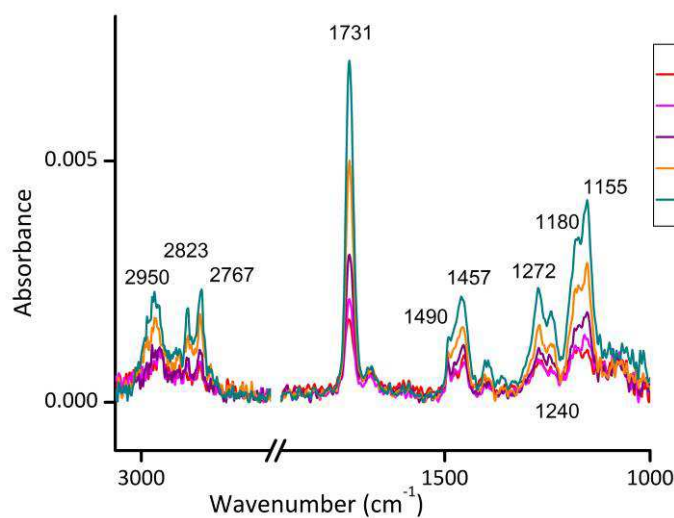


Figure S2. Infrared spectra along the P(DMAEMA) gradient. The positions A-I correspond to the positions indicated in Figure S3. See Table S1 for peak assignments.

Table S1. Peak assignments for the infrared spectra in Figures S1 and S2.

Peak position (cm ⁻¹)	Assignment
1184	C–O stretching coupled with O–H in-plane bending
1242/1266	C–C–O stretching
1391	CH ₃ symmetric bending
1447	CH ₂ scissoring
1483	CH ₃ asymmetric bending
1733	C=O stretching
2769/2821	symmetric stretching vibrations of -CH ₃ of the tertiary amine groups
2959	C-H stretch methyl

Interaction forces between a negatively charged probe and a single cationic gradient layer

In an effort to simplify the interpretation of the interaction of the MUA probe with the PDMAEMA gradient, and to clarify the steric contribution to the interaction, a single PDMAEMA gradient was investigated using an MUA probe without interference from a PMAA bottom layer, see Figure S3.

Figures S3b and S3c show the force curves on the single-layer PDMAEMA gradient probed by an MUA probe in 10 mM PB buffer at pH 6.0. The gradient starts from curve B and the thickness continuously increases up to curve G. In Figure S3b, the attraction reaches a maximum in curve E, where also the attractive interaction starts at the largest separation. For the last two curves, F and G, both the magnitude and the range of the attraction is smaller. This may have different explanations; first, increasing steric repulsion arising from a denser polymer network where the PDMAEMA layer is thicker may reduce the net attraction, but also the fact that the point of zero separation in the data is assumed to be in the constant compliance region may lead to misinterpretation, since a thicker polymer layer trapped between the probe and the surface will shift the apparent zero of separation. From Figure S3c, little quantitative information can be extracted at close separation since the probe jumps to contact from large separation in most of the curves, and thus a separation of steric and electrostatic forces in this region is not possible. For this reason, it was essential to see how an uncharged probe interacts with the hydrogel gradient, in order to understand the role of steric contribution to the total forces, and experiments with a C16OH probe were conducted.

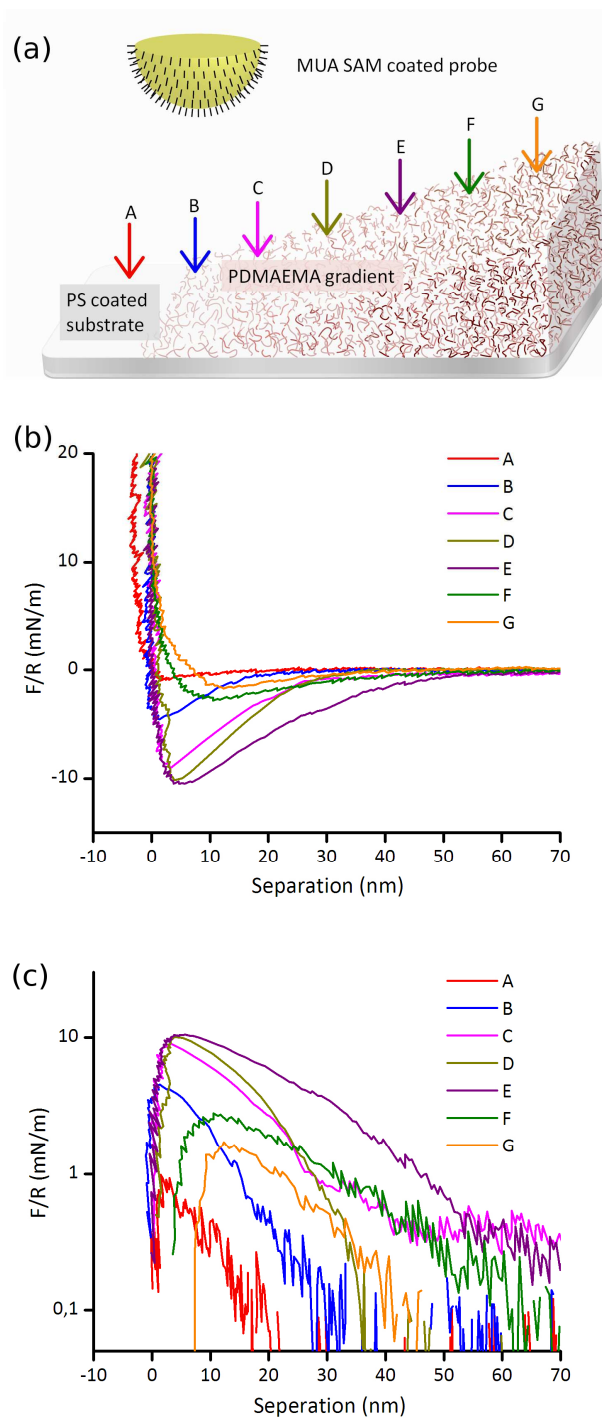


Figure S3. (a) The experiment geometry (not to scale). The PDMAEMA gradient is probed by an MUA SAM-coated probe, and arrows indicate the positions where force curves were obtained. Position A is bare PS, and the PDMAEMA gradient starts from curve B, increasing in thickness towards G. (b) Force curves obtained along a single PDMAEMA gradient in 10 mM PB buffer at pH 6.0. (c) The same data plotted on a log scale (all data plotted with reversed sign).