Supporting Information

Tuning the pH Response of Weak Polyelectrolyte Brushes with Specific Anion Effects

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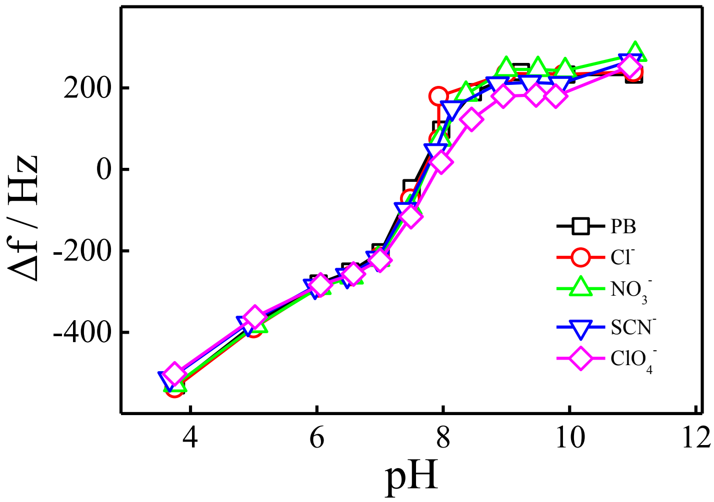

Figure S1. Shift in frequency (Δf) of PDMAEMA brushes as a function of pH in the PB solution and in the presence of different types of anions.

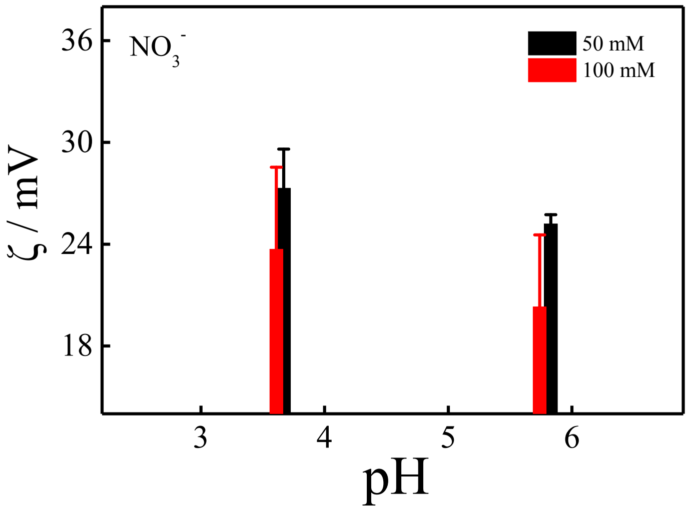

Figure S2. pH dependence of the surface zeta potential (ζ) of PDMAEMA brushes in the presence of NO_3^- at the salt concentrations of 50 and 100 mM.

Here, the surface zeta potential of PDMAEMA brushes in the presence of Cl^- and SCN^- cannot be measured due to the occurrence of electrochemical reaction at the salt concentrations of 50 and 100 mM.[S1,S2] The surface zeta potential of PDMAEMA brushes in the presence of ClO_4^- also cannot be measured at the salt concentrations of 50 and 100 mM due to the strong interactions between the ClO_4^- anions and the positively charged quaternary ammonium groups grafted on the surface of tracer particles.[S3]

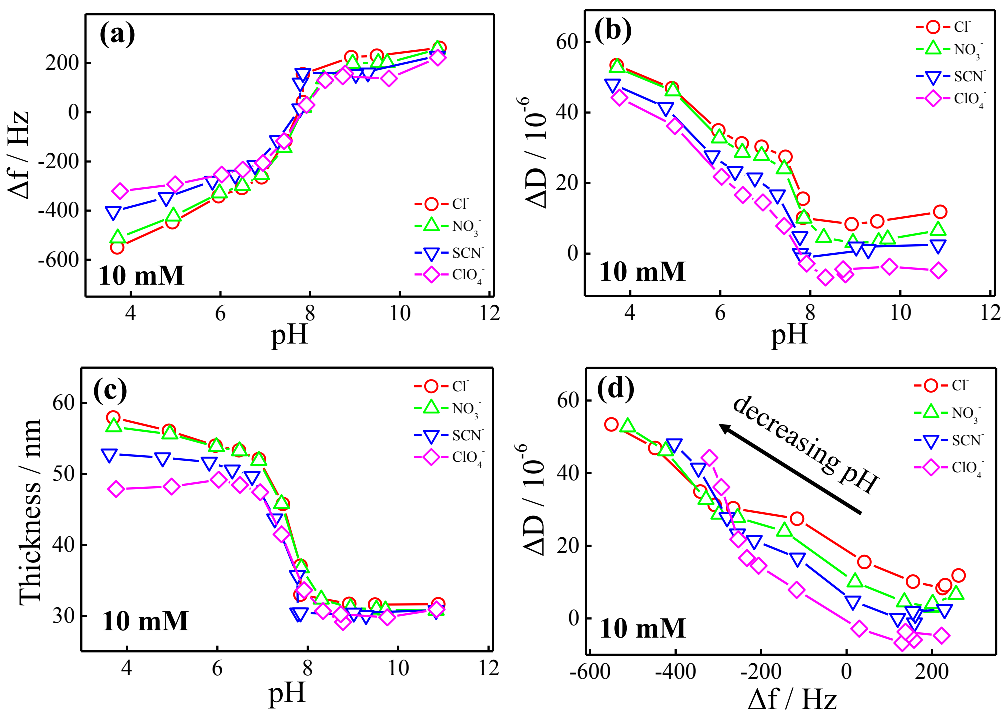

Figure S3. (a) pH dependence of the frequency shift (Δf) of PDMAEMA brushes as a function of anion identity at the salt concentration of 10 mM. (b) pH dependence of the dissipation shift (ΔD) of PDMAEMA brushes as a function of anion identity at the salt concentration of 10 mM. (c) pH dependence of the wet thickness of PDMAEMA brushes as a function of anion identity at the salt concentration of 10 mM. (d) Dissipation shift (ΔD) versus frequency shift (Δf) of the PDMAEMA brushes as a function of anion identity at the salt concentration of 10 mM during the change in pH.

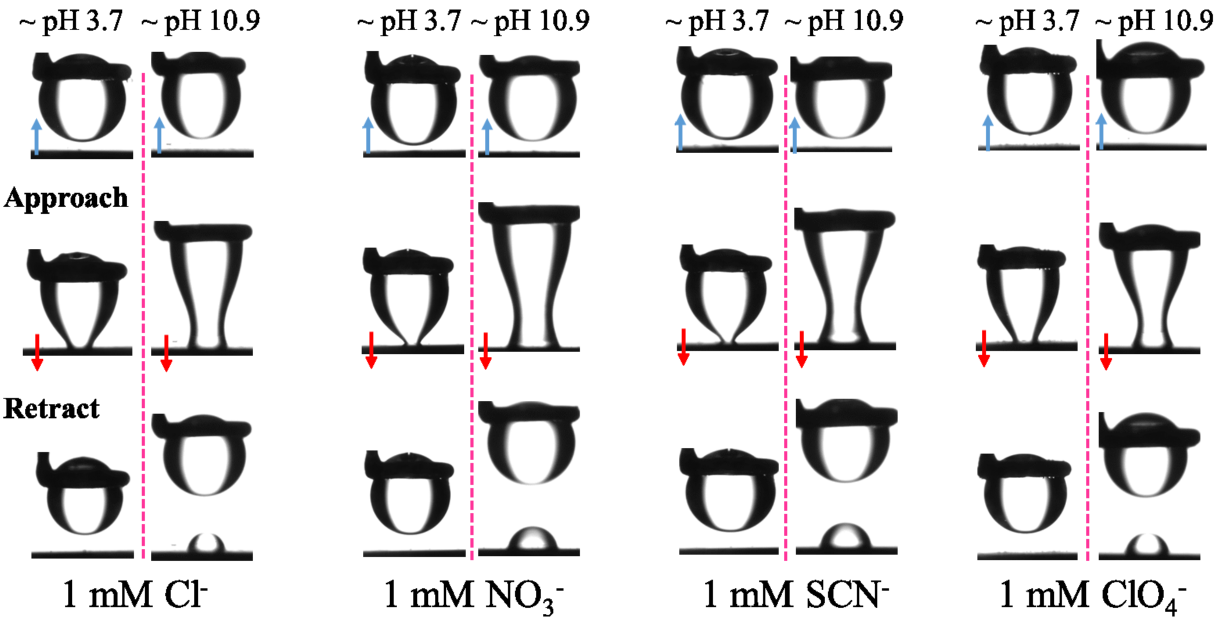

Figure S4. A series of photos taken of the PDMAEMA brushes approaching and retracting from an oil (n-hexadecane) droplet at pH of ~ 3.7 and ~ 10.9 for the different types of anions at a salt concentration of 1 mM during measurements of the adhesive force.

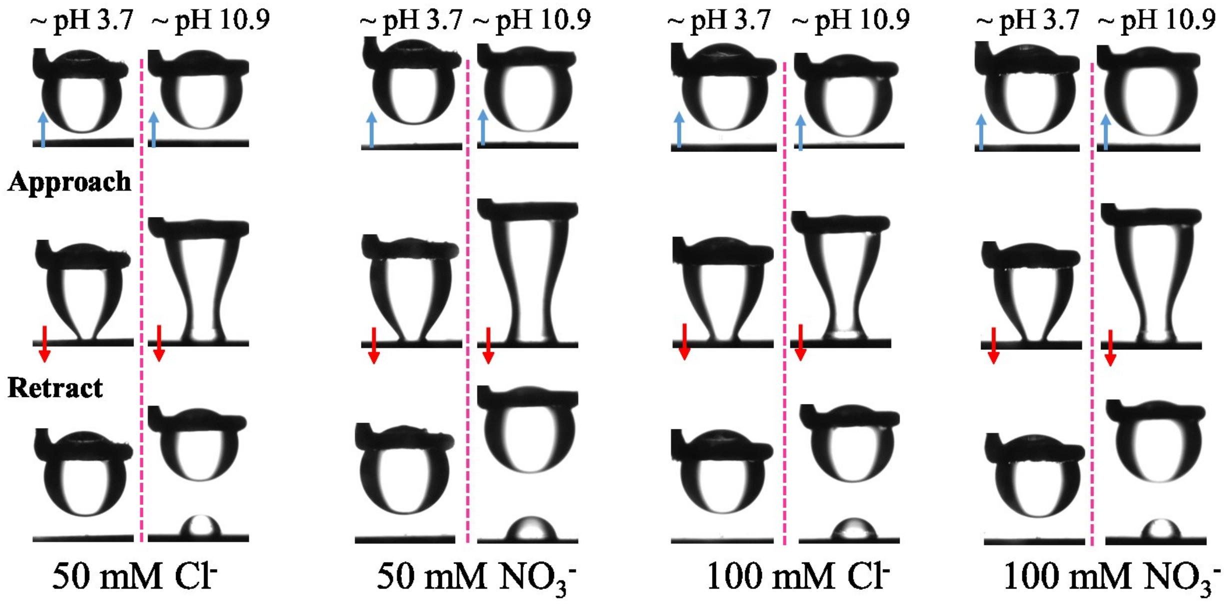

Figure S5. A series of photos taken of the PDMAEMA brushes approaching and retracting from an oil (n-hexadecane) droplet at pH of ~ 3.7 and ~ 10.9 for Cl^- and NO_3^- at the salt concentrations of 50 and 100 mM during measurements of the adhesive force.

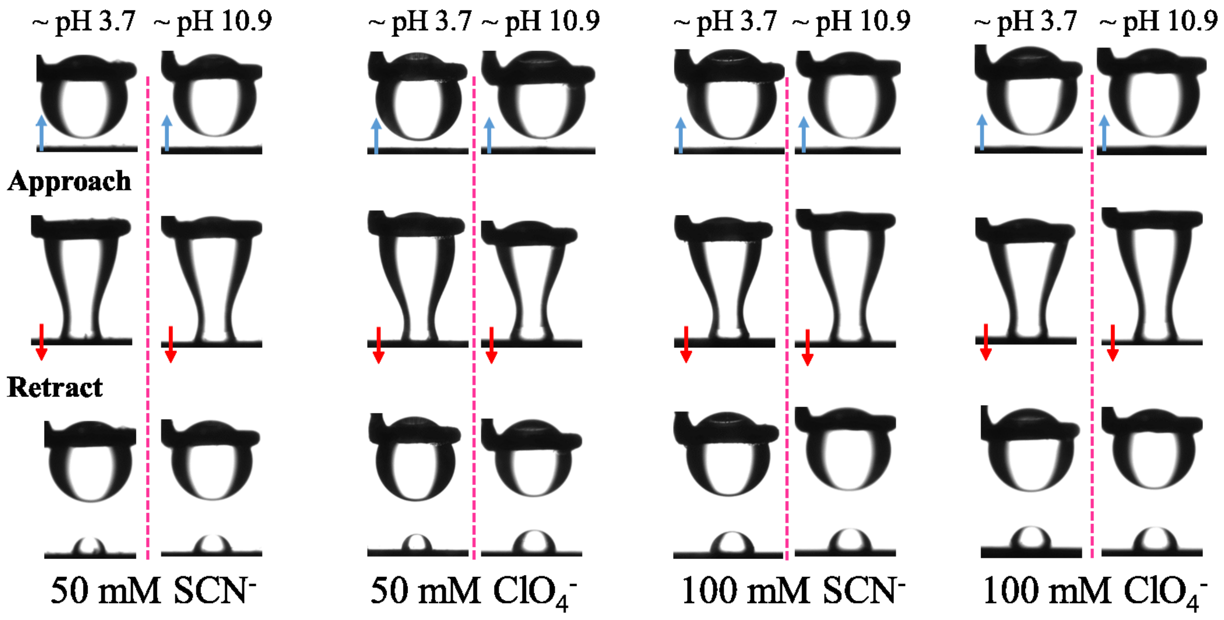

Figure S6. A series of photos taken of the PDMAEMA brushes approaching and retracting from an oil (n-hexadecane) droplet at pH of ~ 3.7 and ~ 10.9 for SCN^- and ClO_4^- at the salt concentrations of 50 and 100 mM during measurements of the adhesive force.

References

S1. Krishnan, P. The Effect of Concentration in Electrochemical Oxidation of Thiocyanate on Platinum Electrode. *J. Solid State Electrochem.* **2007**, *11*, 1327-1334.

S2. Thomas, T. E.; Al Aani, S.; Oatley-Radcliffe, D. L.; Williams, P. M.; Hilal, N. Laser Doppler Electrophoresis and Electro-Osmotic Flow Mapping: A Novel Methodology for the Determination of Membrane Surface Zeta Potential. *J. Membr. Sci.* **2017**, *523*, 524-532.

S3. Kou, R.; Zhang, J.; Chen, Z.; Liu, G. M. Counterion Specificity of Polyelectrolyte Brushes: Role of the Specific Ion- Pairing Interactions. *ChemPhysChem* **2018**, *19*, 1404-1413.