Supporting Information for

Wet Adhesion Strategy Via Synergistic Cation-π and Hydrogen bonding Interactions of Antifouling Zwitterions and Mussel-Inspired Binding Moiety

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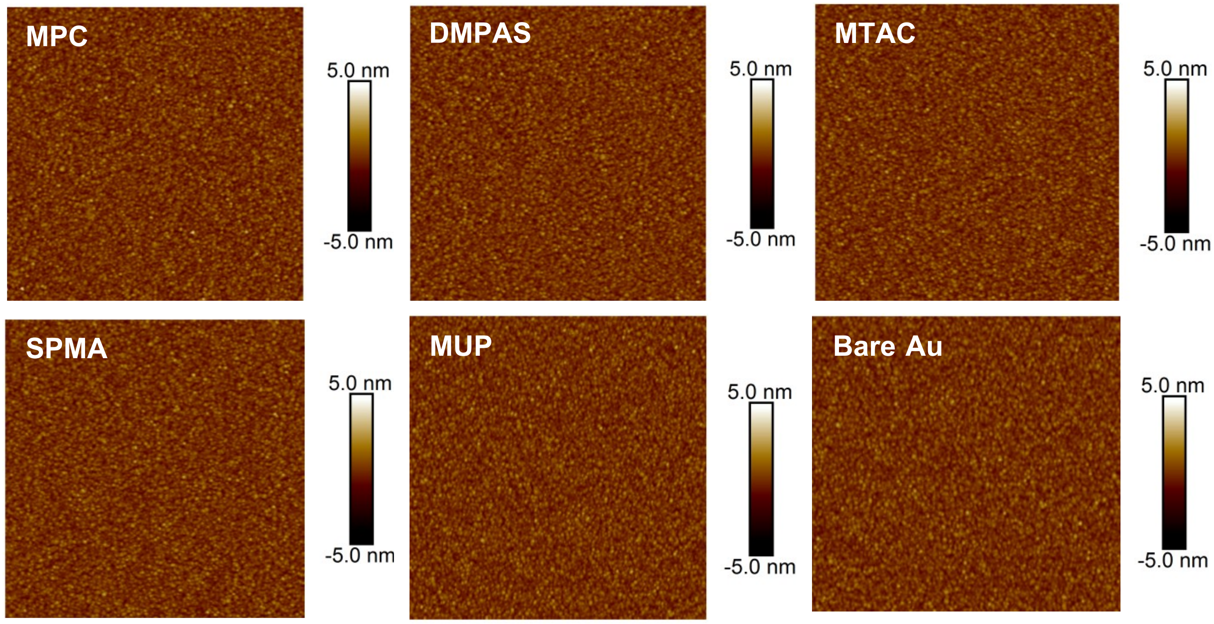


Figure S1. AFM topographic image (5×5 μm²) of MPC, DMAPS, MTAC, SPMA and MUP self-assembled gold surfaces and the bare gold without self-assembly.

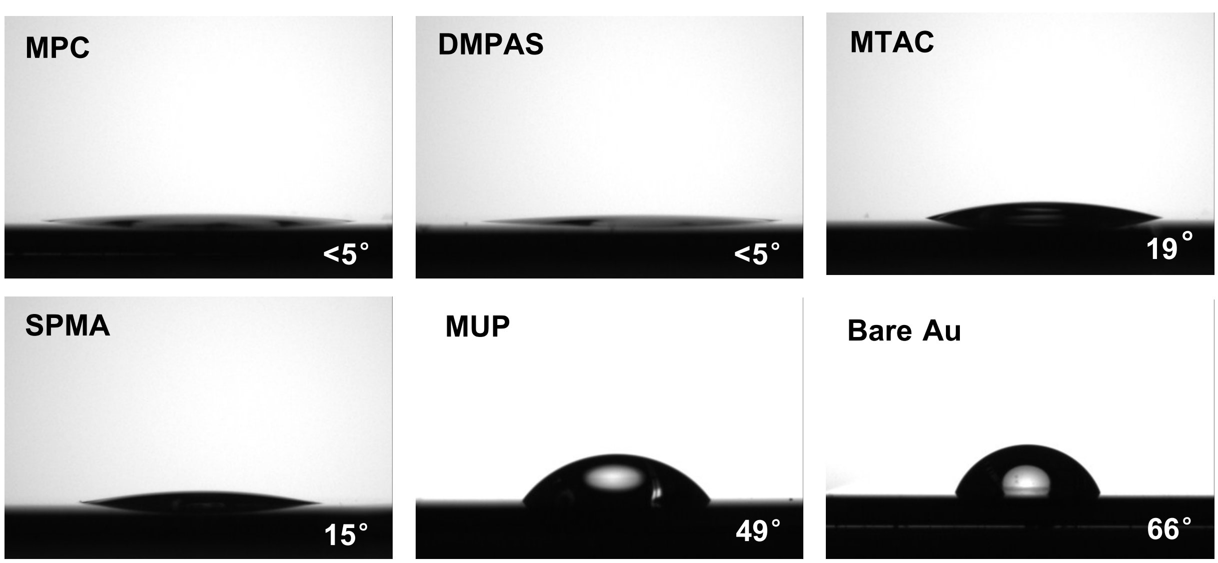


Figure S2. Water contact angle of MPC, DMAPS, MTAC, SPMA and MUP self-assembled gold surfaces and the bare gold without self-assembly.

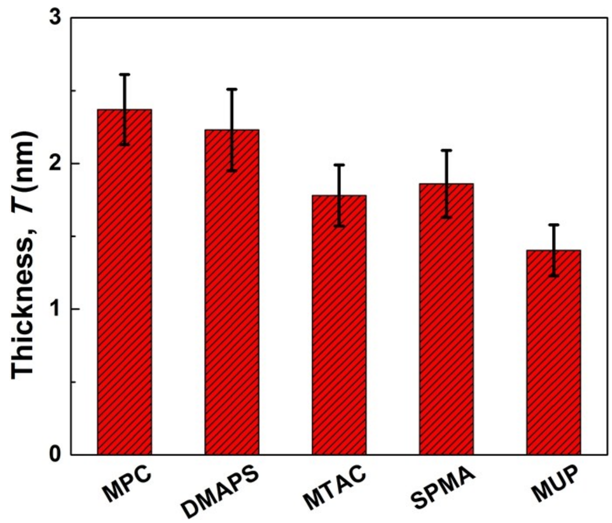


Figure S3. Thickness of self-assembled MPC, DMAPS, MTAC, SPMA and MUP monolayers.

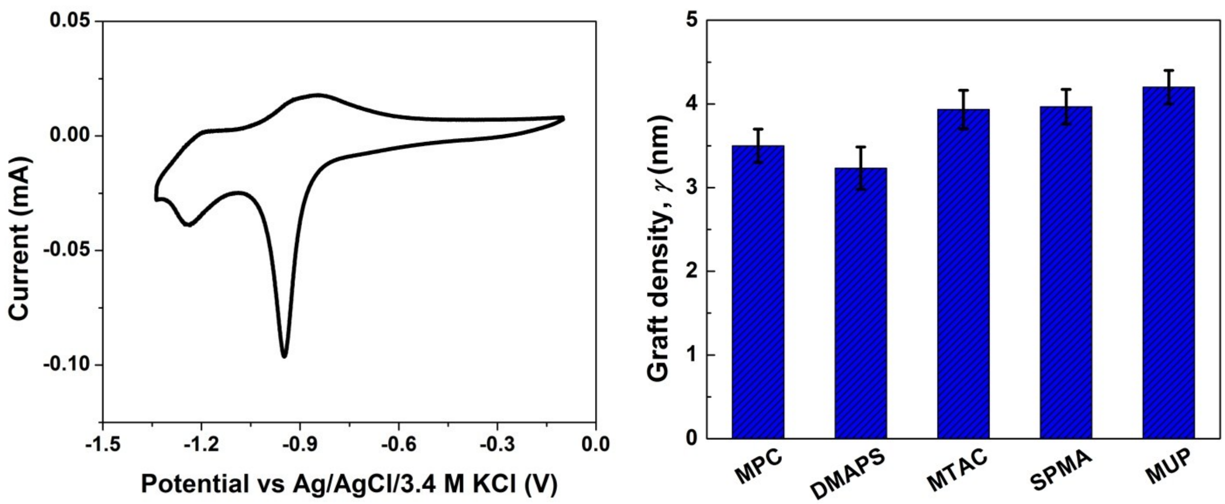
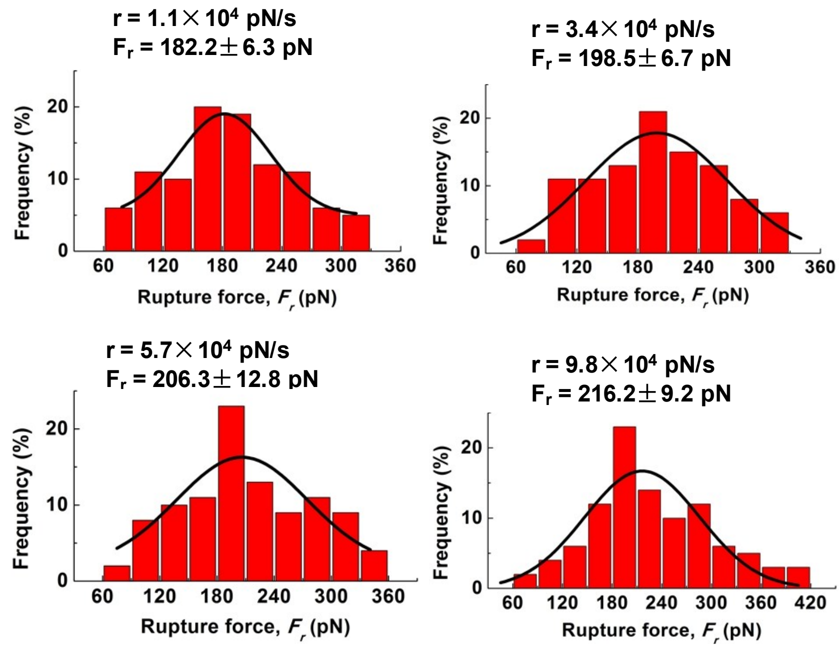
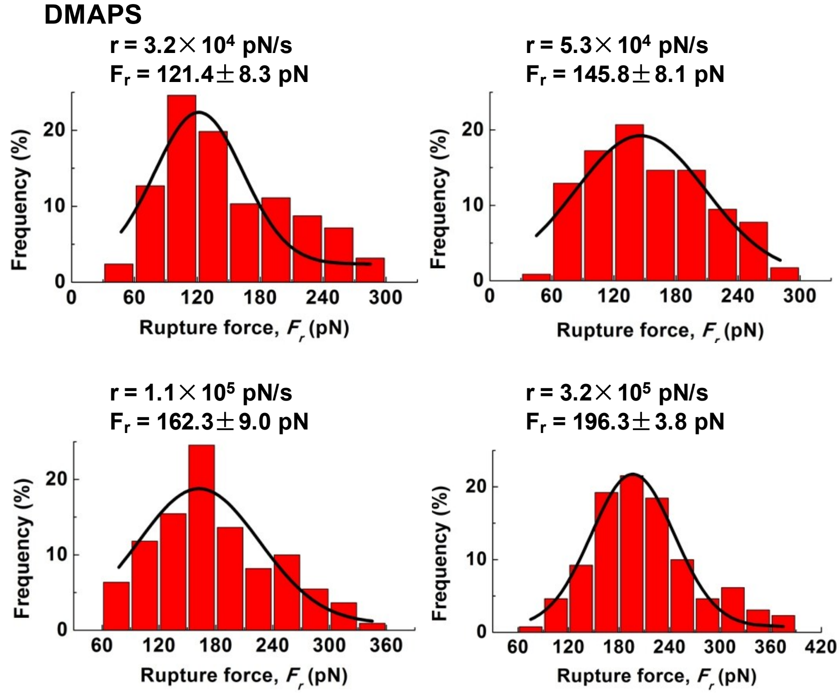


Figure S4. Representative CV curve for self-assembled monolayers with the reduction peak (Left) and the calculated graft density of MPC, DMAPS, MTAC, SPMA and MUP monolayers (Right).

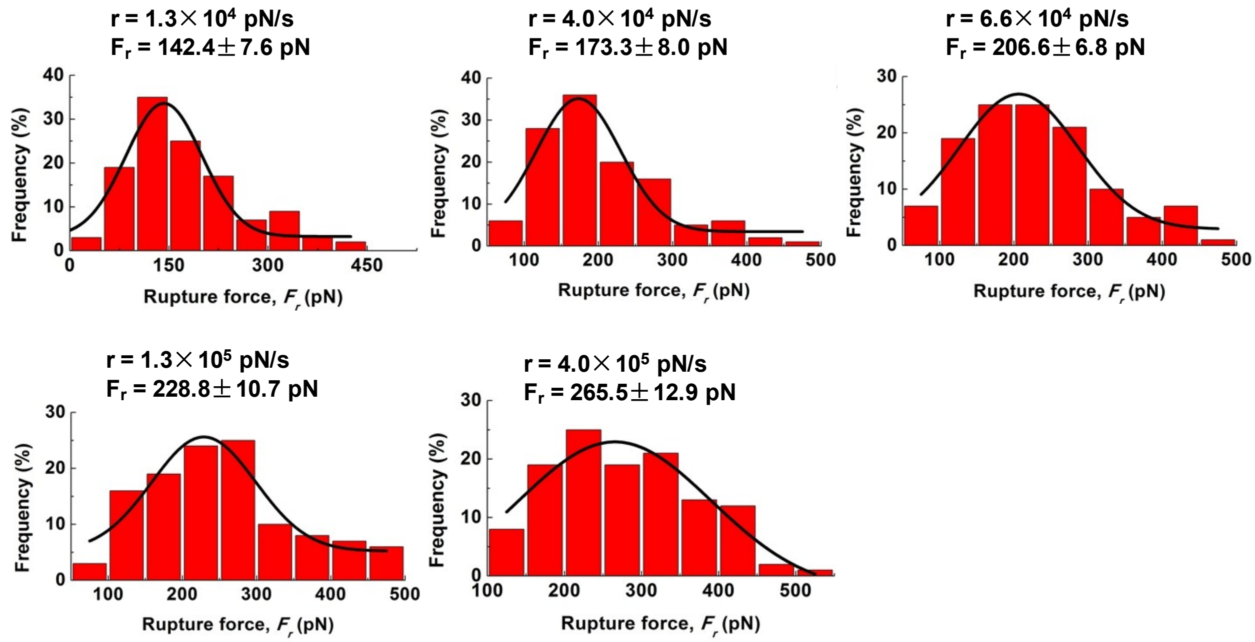
MPC


r = 1.1×10⁴ pN/s
F_r = 182.2±6.3 pN
r = 3.4×10⁴ pN/s
F_r = 198.5±6.7 pN
r = 5.7×10⁴ pN/s
F_r = 206.3±12.8 pN
r = 9.8×10⁴ pN/s
F_r = 216.2±9.2 pN
Frequency (%)
Rupture force, F_r (pN)

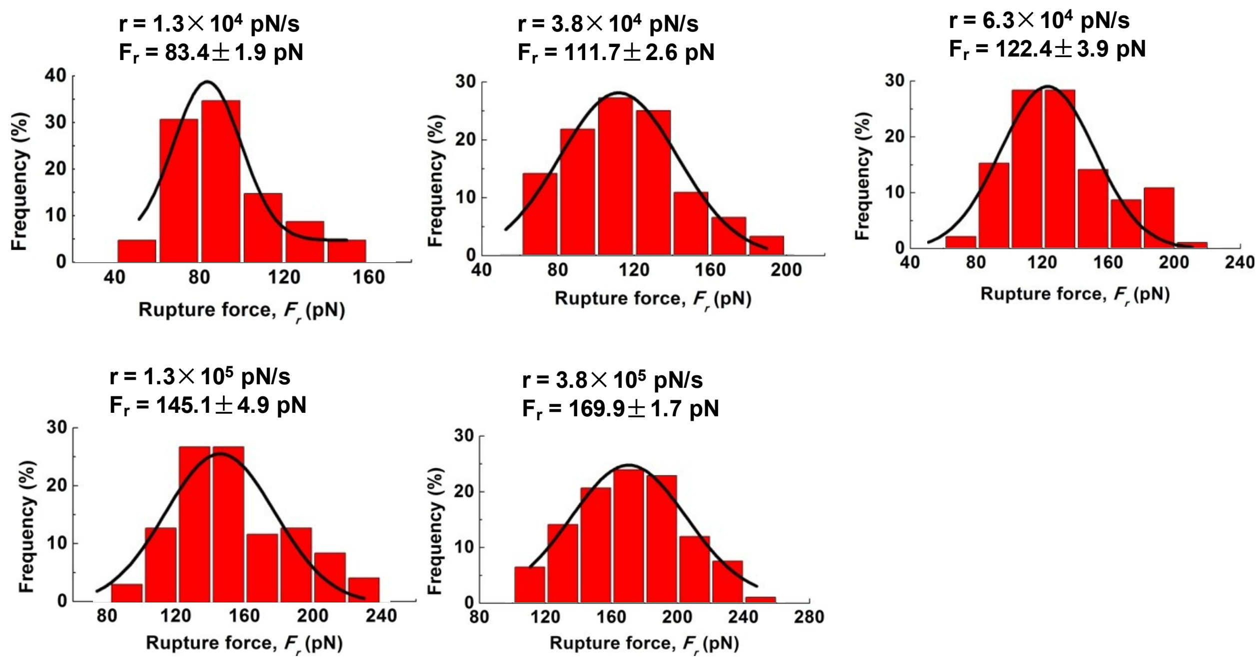
DMAPS


r = 3.2×10⁴ pN/s
F_r = 121.4±8.3 pN
r = 5.3×10⁴ pN/s
F_r = 145.8±8.1 pN
r = 1.1×10⁵ pN/s
F_r = 162.3±9.0 pN
r = 3.2×10⁵ pN/s
F_r = 196.3±3.8 pN
Frequency (%)
Rupture force, F_r (pN)

MTAC


r = 1.3×10^4 pN/s
F_r = 142.4±7.6 pN
r = 4.0×10^4 pN/s
F_r = 173.3±8.0 pN
r = 6.6×10^4 pN/s
F_r = 206.6±6.8 pN
r = 1.3×10^5 pN/s
F_r = 228.8±10.7 pN
r = 4.0×10^5 pN/s
F_r = 265.5±12.9 pN
Frequency (%)
Rupture force, F_r (pN)

SPMA


r = 1.3×10^4 pN/s
F_r = 83.4±1.9 pN
r = 3.8×10^4 pN/s
F_r = 111.7±2.6 pN
r = 6.3×10^4 pN/s
F_r = 122.4±3.9 pN
r = 1.3×10^5 pN/s
F_r = 145.1±4.9 pN
r = 3.8×10^5 pN/s
F_r = 169.9±1.7 pN
Frequency (%)
Rupture force, F_r (pN)

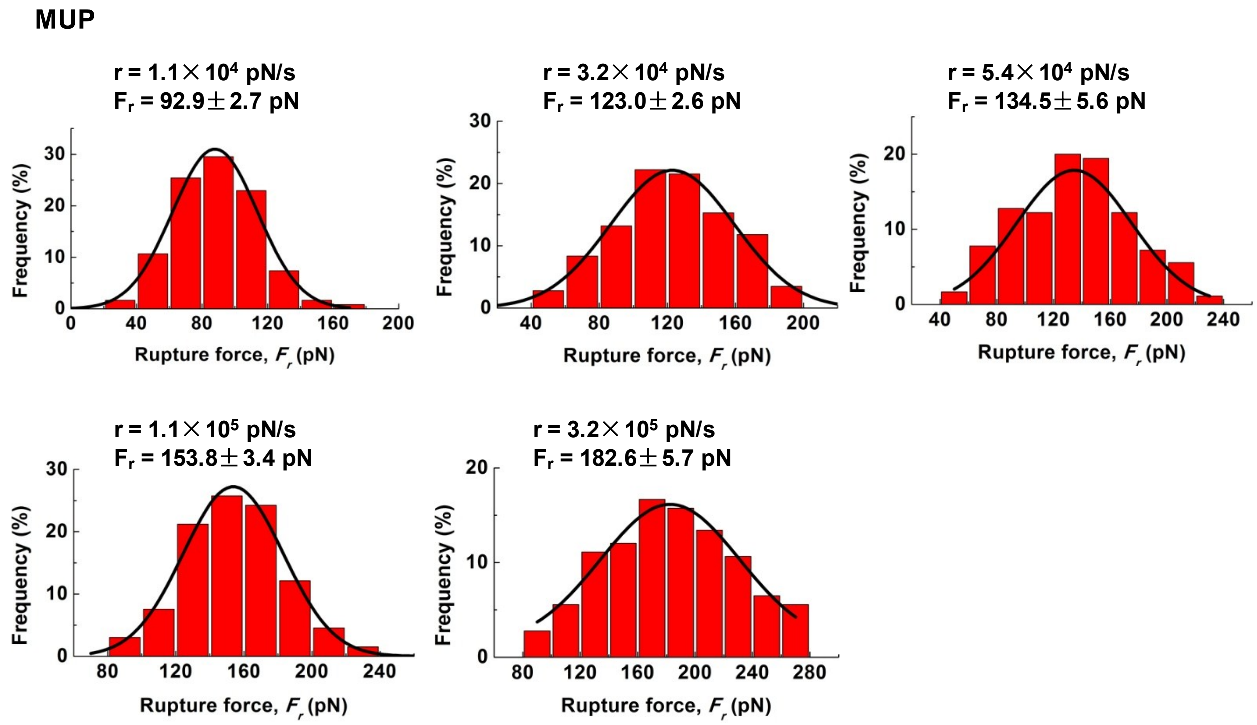


Figure S5. The histogram of rupture forces and the fitted Gaussian distribution for MPC, DMAPS, MTAC, SPMA and MUP.

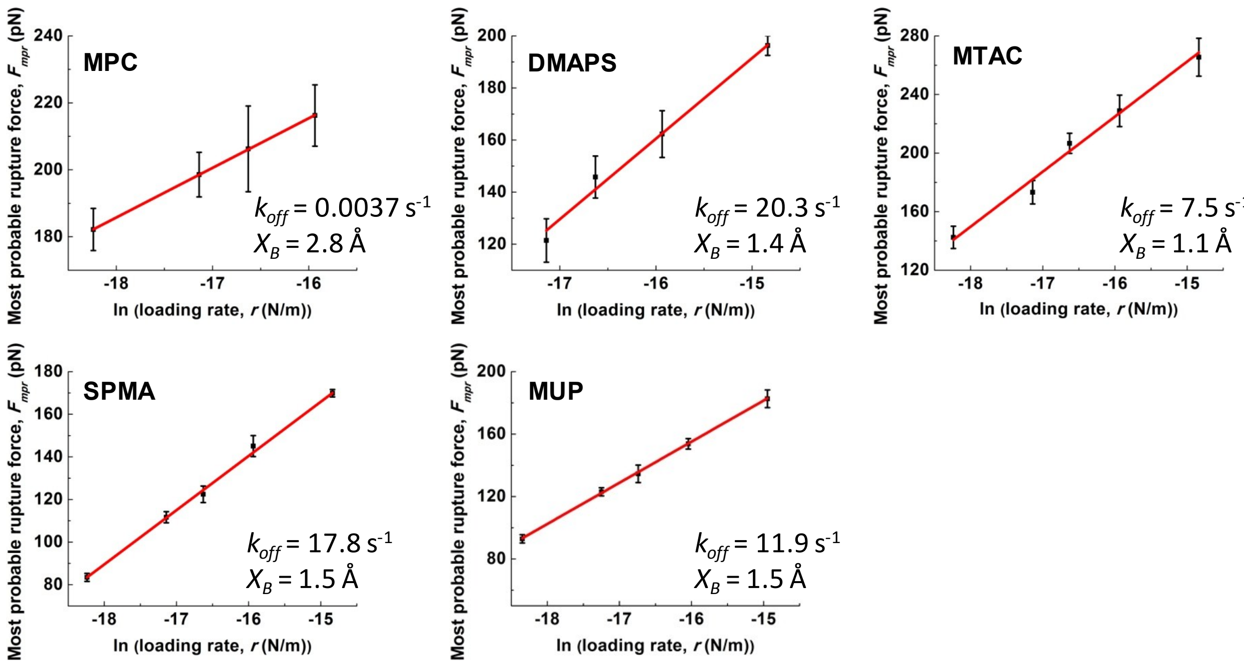


Figure S6. The logarithmic relation of the most probable rupture force F_{mpr} as a function of the loading rate r for MPC, DMAPS, MTAC, SPMA and MUP.

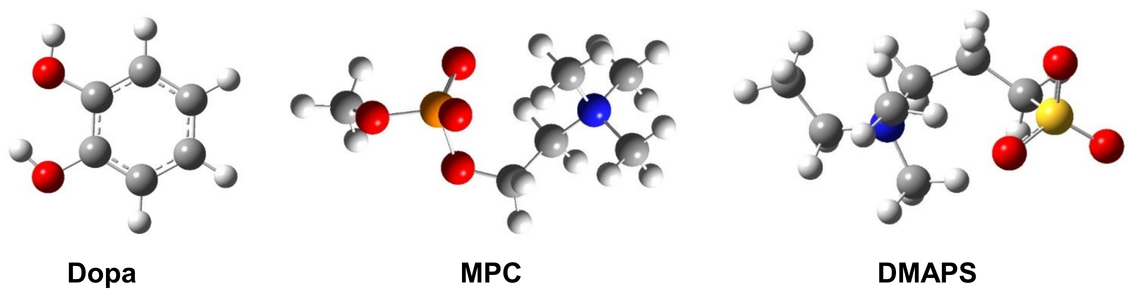


Figure S7. Optimized configurations of the headgroups of Dopa, MPC and DMAPS (grey: carbon; white: hydrogen; red: oxygen; navy: nitrogen; orange: phosphorus; yellow: sulfur).

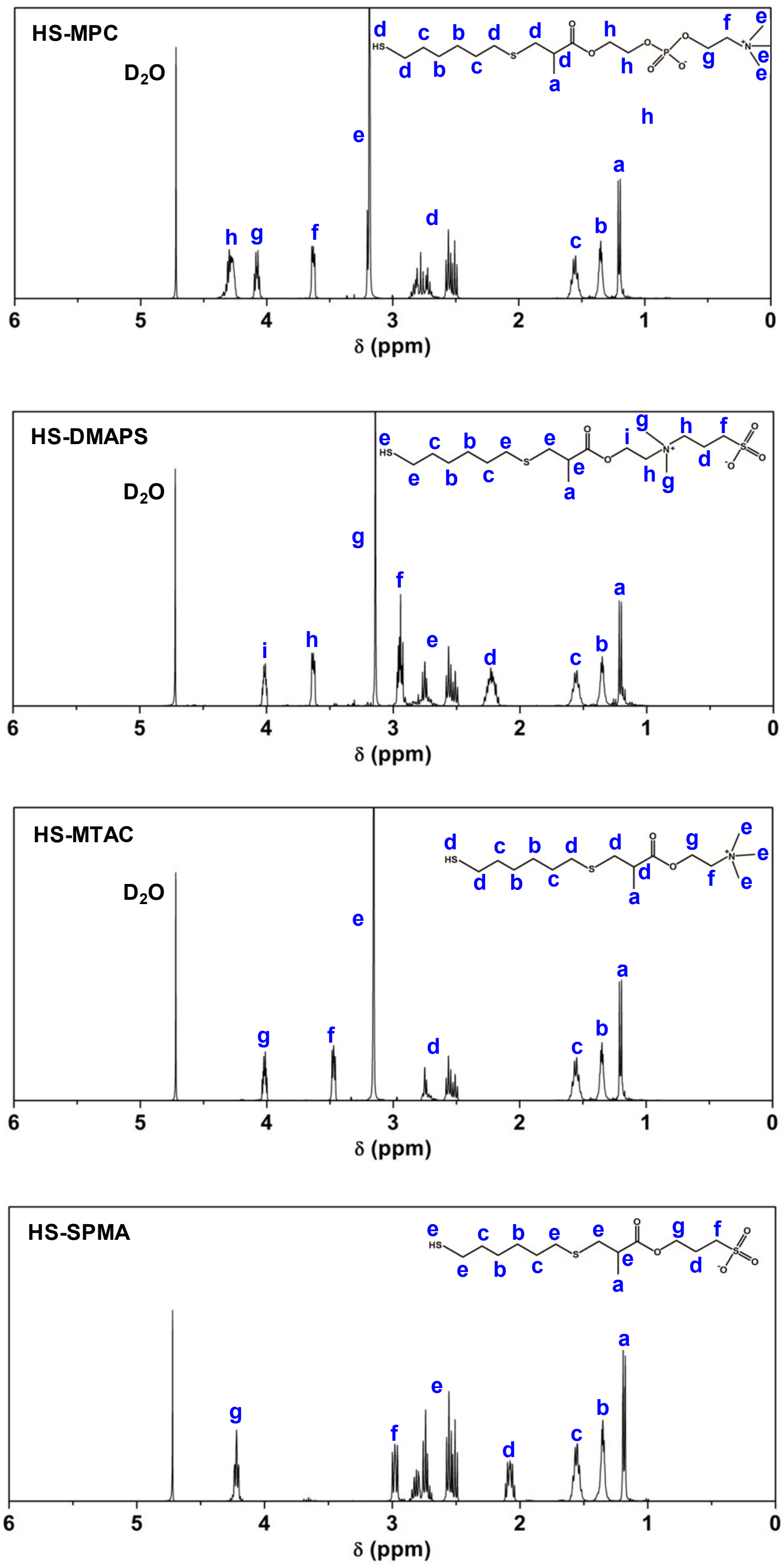


Figure S8. ^{1}H NMR spectra of HS-MPC, HS-DMAPS, HS-MTAC and HS-SPMA.

The preparation and test of polymer adhesive were conducted using the previously reported method.[1-3] Typically, the poly(MPC-co-DMA) solution was evenly applied onto the bond region of two glass substrates. Thereafter, these two glass substrates were pressed with an overlapped area of 2.5 cm × 2.5 cm and then immersed in water for 24 h at room temperature. The strength of the polymer adhesive was measured by pulling apart the glass substrates using the hanged weights in a water bath. It was found that the glass substrates could not be pulled apart under the force of 100 g weight, and the underwater adhesion behavior of the synthesized polymer in this work is comparable to those reported in the literatures.[1-3]

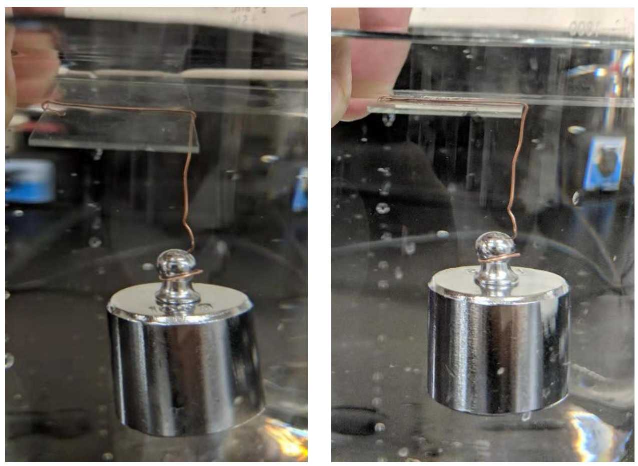


Figure S9. Test of poly(MPC-*co*-DMA) as polymer adhesive in water bath.

References

1. H. Shao and R. J. Stewart, *Advanced materials*, 2010, **22**, 729-733.
2. J. D. White and J. J. Wilker, *Macromolecules*, 2011, **44**, 5085-5088.
3. M. J. Brennan, B. F. Kilbride, J. J. Wilker and J. C. Liu, *Biomaterials*, 2017, **124**, 116-125.