*Suppporting Information for*

# Co-assembled Supramolecular Organohydrogels of Amphiphilic Zwitterion and Polyoxometalate with Controlled Microstructures

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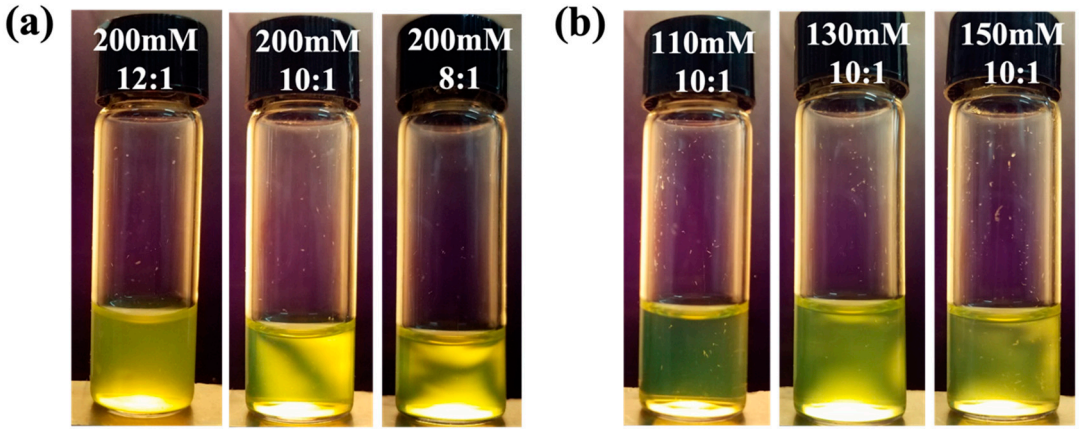


**Figure S1.** Optical photographs of organohydrogels observed with crossed polarizers: (a) samples with different $(C8)_2$MeAS/HPMo molar ratios and fixed $(C8)_2$MeAS concentration of 200 mM; (b) samples with various $(C8)_2$MeAS concentrations and the fixed $(C8)_2$MeAS/HPMo molar ratio of 10:1.

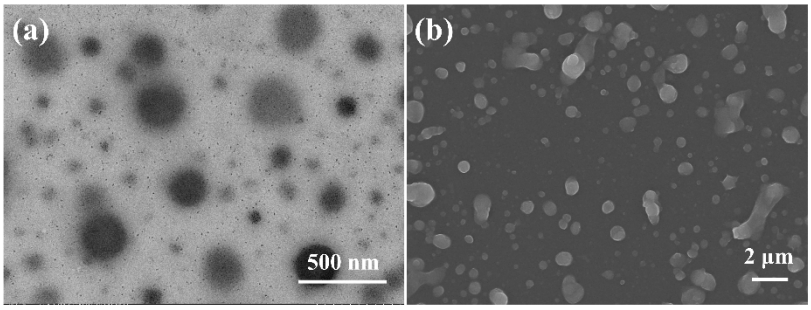


**Figure S2.** TEM (a) and SEM (b) images of the organohydrogel with $(C8)_2$MeAS/HPMo molar ratio of 5:1. The concentration of $(C8)_2$MeAS was fixed at 200 mM.

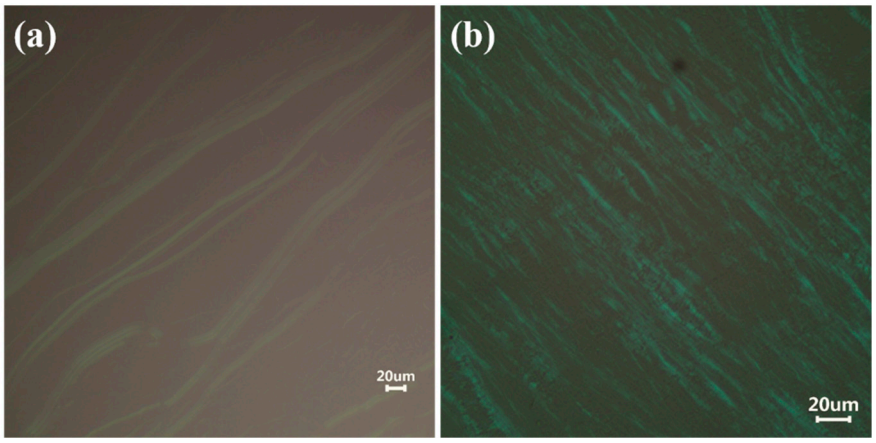


**Figure S3.** POM images of samples with different $(C8)_2$MeAS concentration: (a) 280 mM (b) 300 mM. The $(C8)_2$MeAS/HPMo molar ratio is fixed at 10:1.

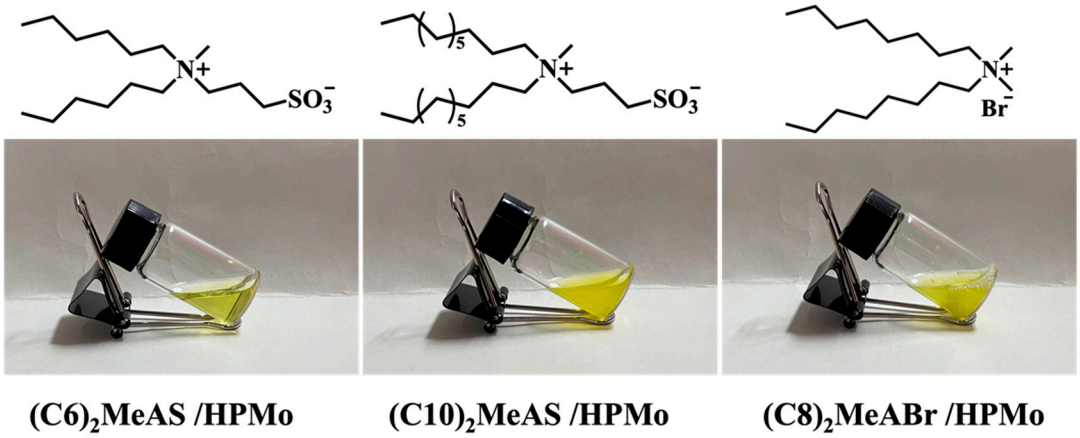


**Figure S4.** Photos of mixtures composed of different amphiphiles and HPMo. The molar ratio between amphiphiles and HPMo was 10:1, and the concentration of amphiphiles was fixed at 200 mM.

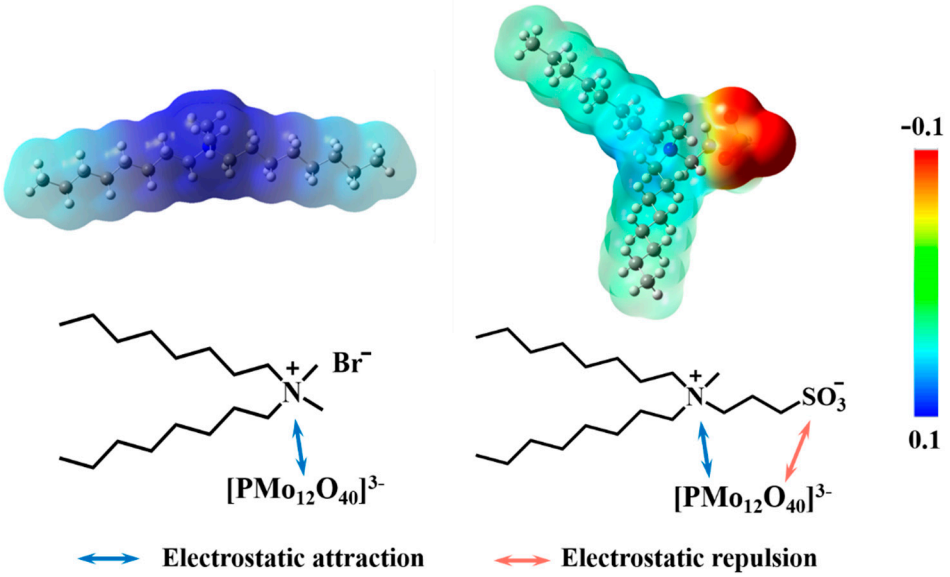


**Figure S5.** Schematic illustration of electrostatic interaction for $(C8)_2$MeABr/HPMo and $(C8)_2$MeAS/HPMo systems.

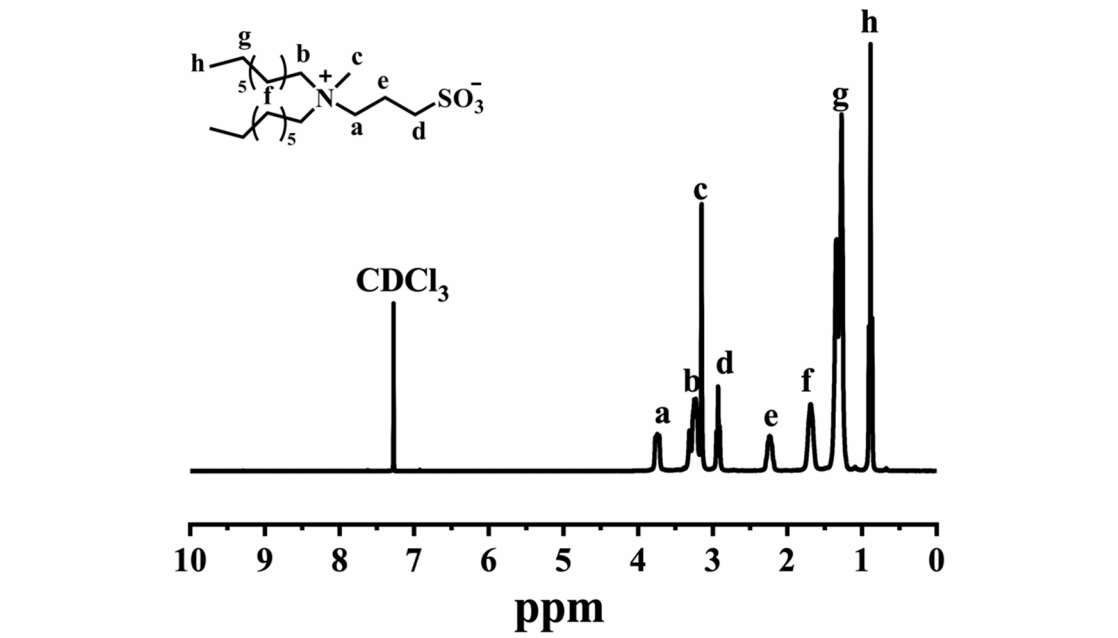


**Figure S6.** The $^{1}$H NMR spectrum of $(C8)_2$MeAS in $CDCl_3$.