Electronic Supplementary Information

Spontaneous vesicle formation and vesicle-to-α-gel transition in aqueous mixtures of sodium monododecylphosphate and guanidinium salts

Meihua Gao, ^a Na Du, * ^a Zhiyin Yao, ^a Ying Li, ^a Nan Chen, ^a Wanguo Hou* ^{a,b}

- ^{*a*} Key Laboratory of Colloid & Interface Chemistry (Ministry of Education), Shandong University, Jinan, 250100, P. R. China.
- ^b National Engineering Technology Research Center of Colloidal Materials, Shandong University, Jinan 250100, P. R. China.



Fig. S1 Photographs of SDP (0.15 wt% or ~5 mM)/H₂O mixtures (a) in the absence of GuSalts and (b–f) in the presence of 0.30 wt% (b) GuCl, (c) GuSO₄, (d) GuSO₃, (e) GuPO₄, amd (f) GuCO₃. All samples were placed at 25.0 °C for at least 48 h before the observation.

To clearly see the sediment, choose black as the background of photos.



Fig. S2 Isotropic phase diagrams of SDP/GuSalt/H₂O ternary systems at 25.0 °C.

Table S1 The pK_a values of acid at 25.0 °C.

| Acid | Chemical formula | $pK_{a1}^{[S1]}$ |
|-------------------|-----------------------------------|------------------|
| Hydrochloric acid | HCl | -6.30 |
| Sulfuric acid | H_2SO_4 | -3.00 |
| Sulfamic acid | NH ₂ SO ₃ H | 0.99 |
| Phosphoric acid | H_3PO_4 | 2.15 |
| Carbonic acid | H_2CO_3 | 6.35 |







Fig. S3 Photographs of SDP (0.15 wt% or ~5 mM)/H₂O mixtures in the presence of (a) 1,1,3,3-tetramethylguanidine, (b) urea, (c) urea hydrogen peroxide, and (d) aminourea hydrochloride. All samples were placed at 25.0 °C for at least 48 h before the observation.



Fig. S4 (a, b) Cryo-TEM images and (c, d) size distributions of SDP/GuCl/H₂O systems with different compositions at 25.0 °C. (a, c) 0.90 wt% (or ~30 mM) SDP, 0.55 wt% (or ~57 mM) GuCl; (b, d) 1.50 wt% (or ~50 mM) SDP, 0.55 wt% (or ~57 mM) GuCl.



Fig. S5 (a, b, e) NS-TEM images and (c, d) size distributions of SDP/GuSO₄/H₂O systems with different compositions at 25.0 °C. (a, c) 0.15 wt% (or ~5 mM) SDP, 0.30 wt% (or ~28 mM) GuSO₄; (b, d) 0.60 wt% (or ~20 mM) SDP, 0.55 wt% (or ~51 mM) GuSO₄; (e) 0.15 wt% (or ~5 mM) SDP, 0.34 wt% (or ~31 mM) GuSO₄.



Fig. S6 (a, b, e) NS-TEM images and (c, d) size distributions of SDP/GuSO₃/H₂O systems with different compositions at 25.0 °C. (a, c) 0.15 wt% (or ~5 mM) SDP, 0.30 wt% (or ~19 mM) GuSO₃; (b, d) 0.60 wt% (or ~20 mM) SDP, 0.55 wt% (or ~35 mM) GuSO₃; (e) 0.15 wt% (or ~5 mM) SDP, 0.49 wt% (or ~31 mM) GuSO₃.



Fig. S7 (a, b, e) NS-TEM images and (c, d) size distributions of SDP/GuPO₄/H₂O systems with different compositions at 25.0 °C. (a, c) 0.15 wt% (or ~5 mM) SDP, 0.30 wt% (or ~28 mM) GuPO₄; (b, d) 0.60 wt% (or ~20 mM) SDP, 0.55 wt% (or ~51 mM) GuPO₄; (e) 0.15 wt% (or ~5 mM) SDP, 0.34 wt% (or ~31 mM) GuPO₄.



Fig. S8 (a, b, e) NS-TEM images and (c, d) size distributions of SDP/GuCO₃/H₂O systems with different compositions at 25.0 °C. (a, c) 0.15 wt% (or ~5 mM) SDP, 0.30 wt% (or ~34 mM) GuCO₃; (b, d) 0.60 wt% (or ~20 mM) SDP, 0.21 wt% (or ~23 mM) GuCO₃; (e) 0.15 wt% (or ~5 mM) SDP, 0.27 wt% (or ~31 mM) GuCO₃.



Fig. S9 ¹H NMR spectra of SDP, CuCl, and SDP/GuCl vesicle in D₂O.



Fig. S10 Energy-minimized structures of sodium monododecylphosphate and guanidine via the density functional theory by Gaussian 9.0. The atom coloring scheme is: C, gray; H, white; P, orange; O, red; N, blue.



Fig. S11 Fluorescence intensity decay of DPH in SDP/GuSalt/H₂O solutions (0.15 wt% or ~5 mM SDP, 0.30 wt% GuSalt) at 25.0 °C ($\lambda_{ex} = 377$ nm, $\lambda_{em} = 450$ nm).

| GuSalt | $\tau_{\rm f} ({\rm ns})$ | χ^2 | $\tau_{\rm R}({\rm ns})$ |
|-------------------|----------------------------|----------|--------------------------|
| GuCl | 9.66, 2.49 | 1.02 | 3.69 |
| GuSO ₄ | 9.62, 2.29 | 1.01 | 3.18 |
| GuSO ₃ | 9.82, 2.62 | 0.98 | 3.25 |
| GuPO ₄ | 9.76, 2.46 | 0.95 | 3.23 |
| GuCO ₃ | 9.83, 2.27 | 0.95 | 2.79 |

Table S2 Lifetime (τ_f) , χ^2 , and correlation time (τ_R) of DPH in SDP/GuSalt/H₂O solutions (0.15 wt% or ~5 mM SDP, 0.30 wt% GuSalt) at 25.0 °C.



Fig. S12 DSC trace for SDP/GuSalt/H₂O systems (~5 mM or 0.15 wt% SDP, 0.30 wt% GuSalt).



Fig. S13 Change of T_r with SDP concentration (*C*) for SDP/GuSalt/H₂O solutions at 8.0 °C.



Fig. S14 Polarizing micrographs of SDP/GuSalt/H₂O systems (0.90 wt% SDP, 0.55 wt% GuSalt) at 8.0 °C. (a) GuSO₄, (b) GuSO₃, and (c) GuPO₄.



Fig. S15 Cryo-TEM images of SDP/GuCl/H₂O system (0.90 wt% SDP, 0.55 wt% GuCl) at 8.0 $^{\circ}$ C.



Fig. S16 Cryo-TEM images of SDP/GuPO₄/H₂O system (0.90 wt% SDP, 0.55 wt% GuPO₄) at 8.0 °C.



Fig. S17 Small-angle X-ray scattering curve of SDP/GuCl/H₂O systems (0.90 wt% SDP, 0.55 wt% GuCl) at 8.0 °C. The scattering peaks for the system can be indexed as a lamellar structure.



Fig. S18 (a) Stress sweep and (b) frequency sweep of gel samples for SDP/GuSalt/H₂O systems (0.90 wt% SDP, 0.55 wt% GuSalt) at 8.0 °C.

Reference

[S1] G. S. James, Lange's Handbook of Chemistry 16th: McGraw-Hill, 2005.