Electronic Supplementary Material (ESI) for Soft Matter

Hexagonal Closest-Packed Spheres Liquid Crystalline Phases Stabilised by Strongly Hydrated Counterions

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Supplementary Material

1. Polarising Optical Micrographs of concentration gradient (surfactant penetration or flooding) samples of cationic surfactants. The images are presented in grey scale to highlight the presence of two distinct isotropic liquid crystal phases seen as bands in the majority of samples (see arrows). The HCPS phase (*$P6_3/mmc$*) is adjacent to the L_1 (micellar) liquid, and the I_1 cubic (*Pn3m*) adjacent to the hexagonal phase.

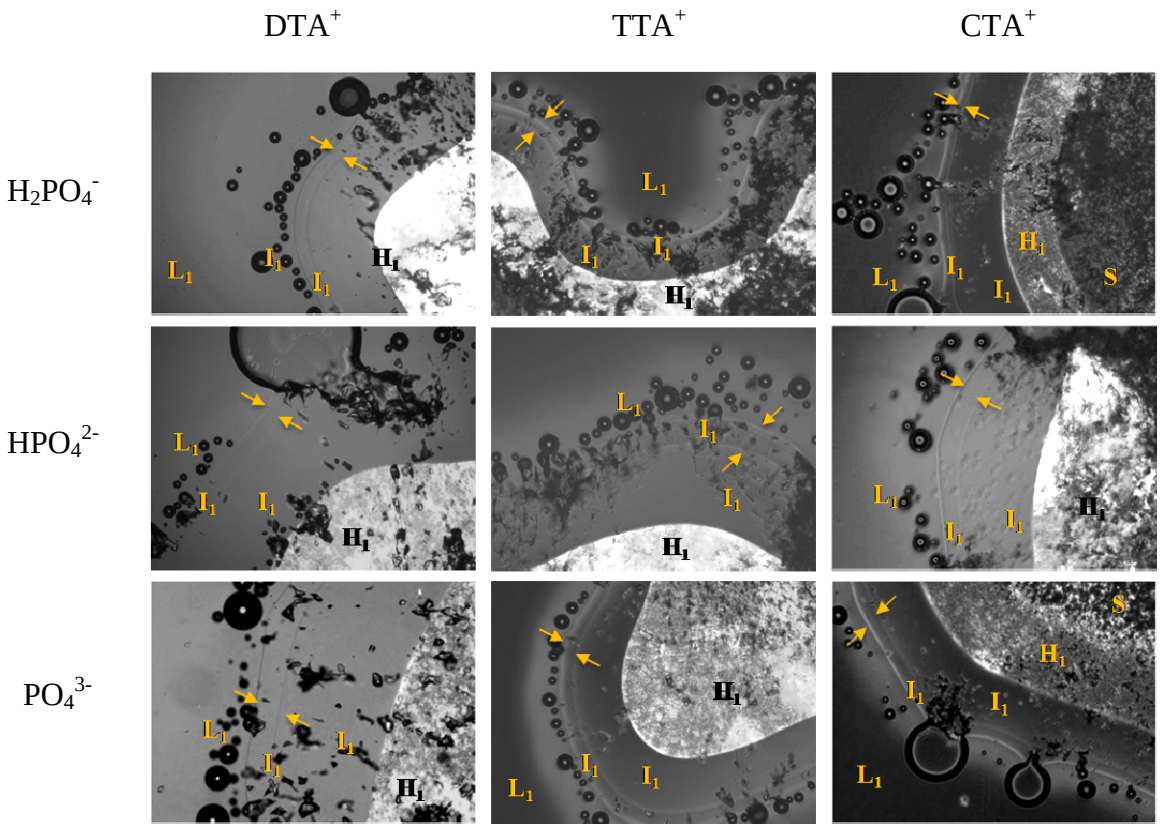

Figure S.1: Polarising optical microscopy images (10×) for alkyltrimethylammonium phosphate surfactants with C_{12} (DTA^+), C_{14} (TTA^+) and C_{16} (CTA^+) alkyl chains.

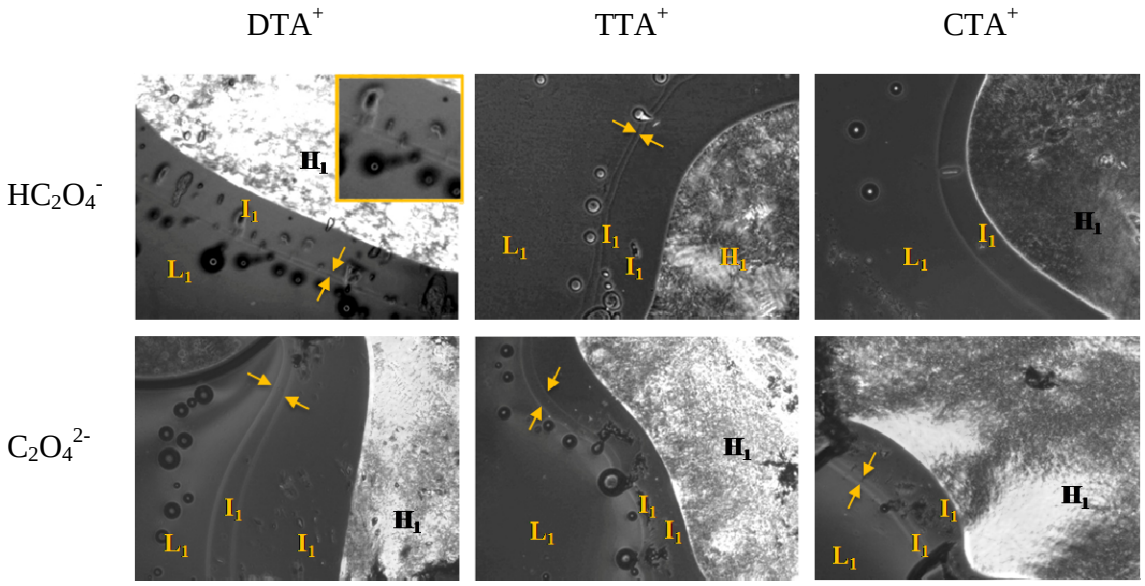

Figure S.2: Polarising optical microscopy images (10×) for alkyltrimethylammonium oxalate surfactants with C_{12} (DTA^+), C_{14} (TTA^+) and C_{16} (CTA^+) alkyl chains.

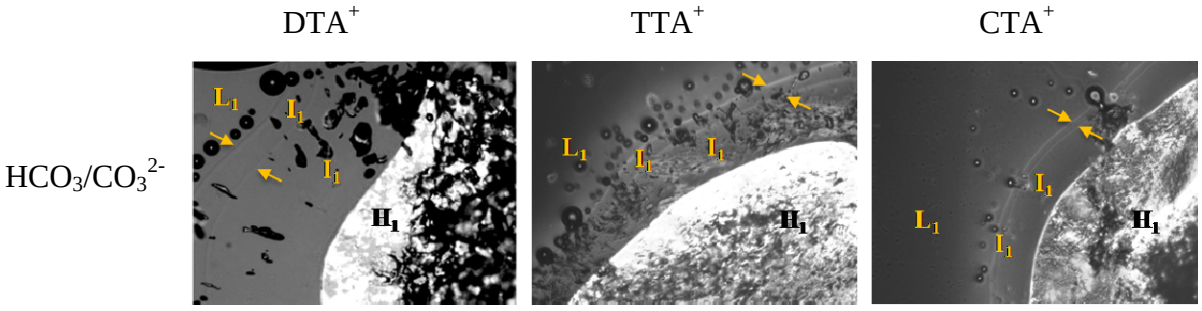

Figure S.3: Polarising optical microscopy images (10×) for alkyltrimethylammonium carbonate surfactants with C_{12} (DTA^+), C_{14} (TTA^+) and C_{16} (CTA^+) alkyl chains.

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2. SAXS patterns from beam overlap on adjacent phases. Figure S.4 shows SAXS pattern of a TTA_2CO_3 sample in which part of the beam was incident on the micellar phase, shown by the broad scattering peak, and part on the adjacent *P6₃/mmc* space group or HCPS phase, for which some of the most intense peaks can be indexed. Interpreted together with the polarising optical microscopy image presented in Figure S.3, the HCPS phase is formed adjacent to the micellar solution (L_1) and the second discrete micellar isotropic phase. The hexagonal phase H_1 lies at still higher concentration.

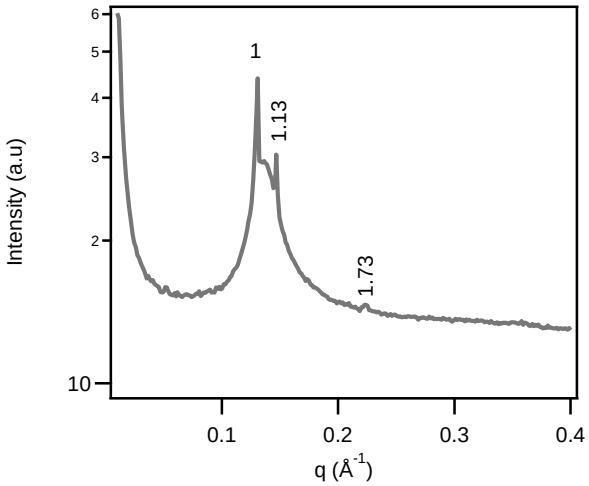

Figure S.4: SAXS pattern from a region within a concentration gradient capillary of TTA_2CO_3 surfactant containing micelles in solution as indicated by the broad peak together with the HCPS phase as indicated by the sharp Bragg diffraction peaks. Assigned numbers indicate the some of the peak ratios observed for the *P6₃/mmc* space group.