

Interplay of formation kinetics for highly oriented and mesostructured silicate-surfactant films at air-water interface

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1. TEM result of a mesostructured Si-CTAB film formed at the air-water interface

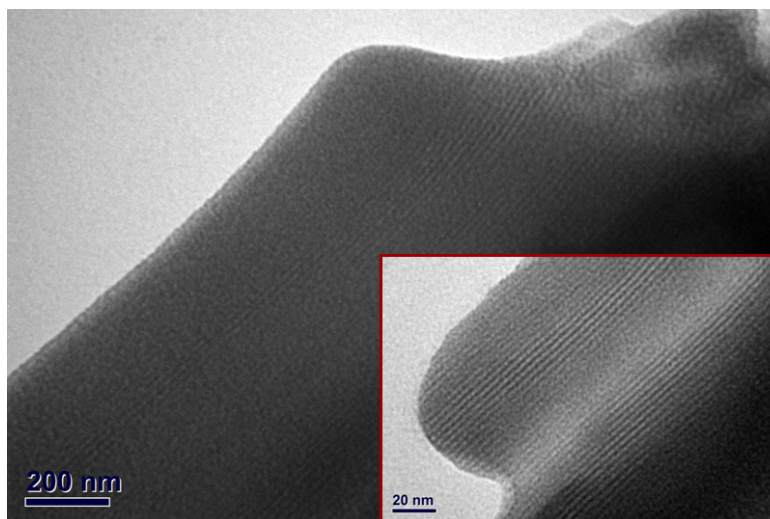


Figure S1. A represented TEM image and a room-in view (inset) for a mesostructured Si-CTAB film harvested from the air-water interface, showing the well-aligned channel structure.

2. CTAB micelles in solution and Si-CTAB micelles at the air-water interface

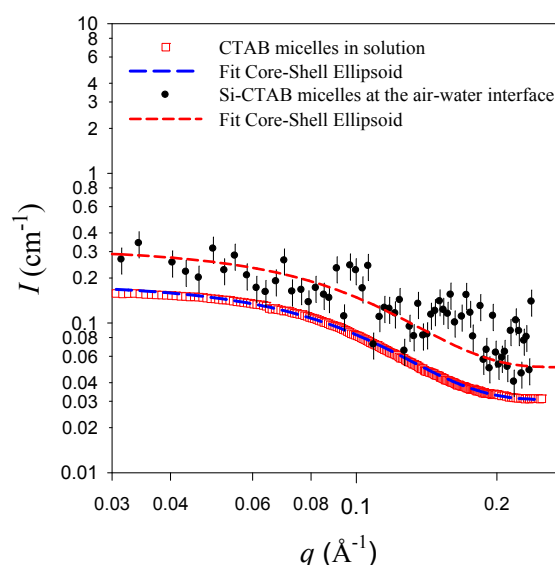


Figure S2. SAXS data for the CTAB solution in the absence of TEOS are fitted (long-dashed curve) using a model of core-shell prolate ellipsoids with semi-major and semi-minor axes of $A = 33.0 \text{ \AA}$ and $B = 17.8 \text{ \AA}$, along with shell thickness (headgroup region) $\sigma = 2.0 \text{ \AA}$. The first nontrivial set of GISAXS data at $t \approx 1300 \text{ s}$ (cf. Figure 1) are fitted (short-dashed curve) using the same model, with resembling parameters of $A = 33.1 \text{ \AA}$, $B = 17.7 \text{ \AA}$, and $\sigma = 2.5 \text{ \AA}$.

3. Silicate concentration effect

Shown in Figure S2 are the phase evolutions of the mesostructure silicate-CTAB films observed at the air-water interface, at 25°C , for the standard solution (at TEOS concentration C_T) and solutions with four-times (i.e., $4C_T$) or one-fourth (i.e., $C_T/4$) of the standard TEOS content. With decreasing silicate concentration from $4C_T$ to $C_T/4$, the induction time was significantly lengthened from 60 to 9000 s, respectively. Nevertheless, the same L-to-H kinetic pathway is followed in all three cases, as illustrated in Figure S2.

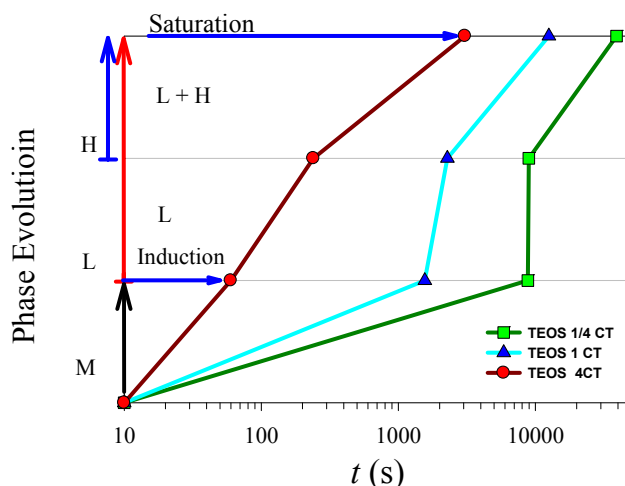


Figure S3. Phase evolution of the three solutions with different TEOS concentrations ($4C_T$, C_T , and $C_T/4$) all followed the same kinetic pathway of isotropic micelles (M) to lamellar (L) phase, and then quantitative transformation of L domains to 2D hexagonal (H) phase.

4. Acidity effects on Si-CTAB micelles

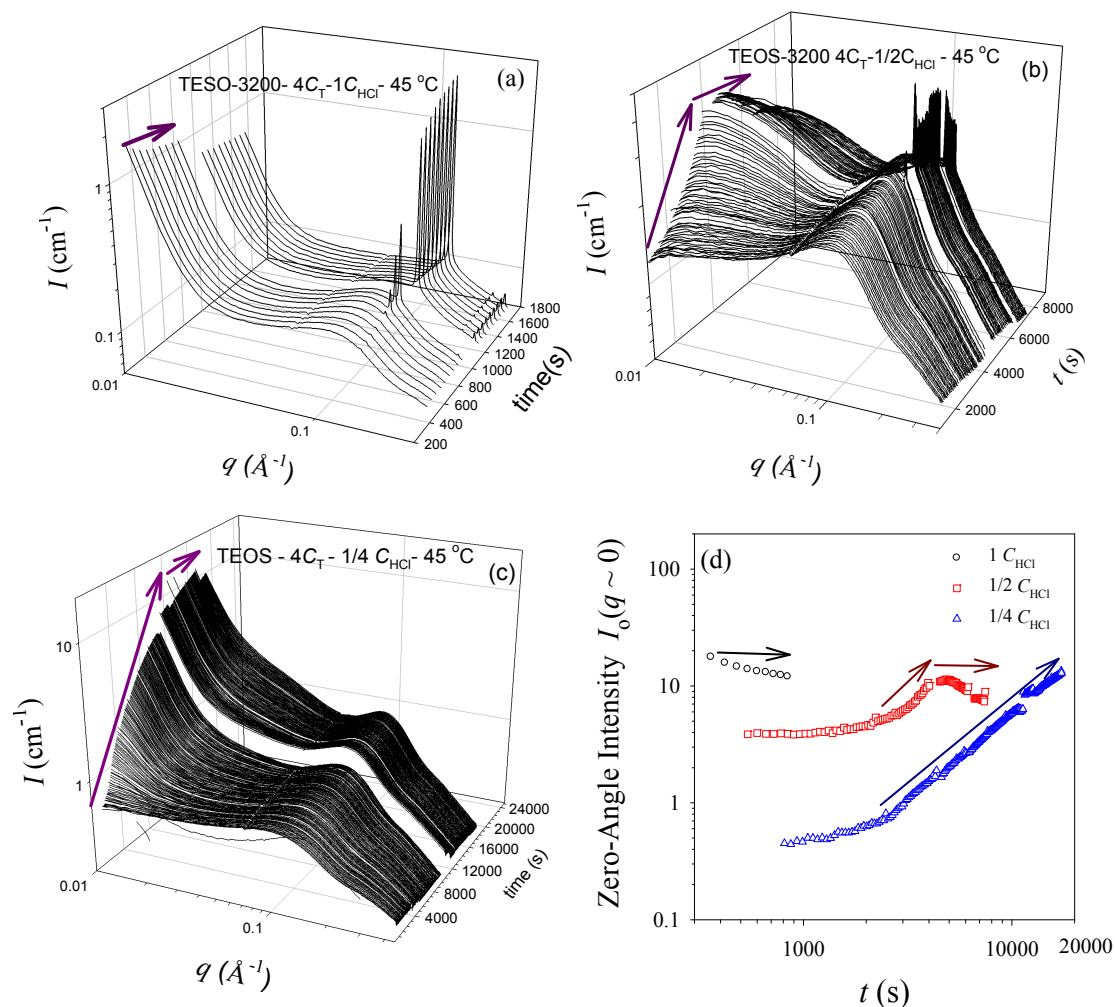


Figure S4: Time-resolved SAXS data measured at 45 °C for (a) a solution of silicate concentrations 4C_T and for similar solutions with decreased acid concentrations of (b) C_{HCl}/2 and (c) C_{HCl}/4. Thick arrows indicate the increasingly slower development of the “zero-angle” intensity I_0 (at $q \approx 0.01$ Å⁻¹) with decreasing acidic level. (d) Corresponding evolutions of I_0 . Note the extremely lengthened growth time for the solution of C_{HCl}/4.

5. In-situ SAXS data for silicate-CTAB sample solutions sealed without the air-water interface

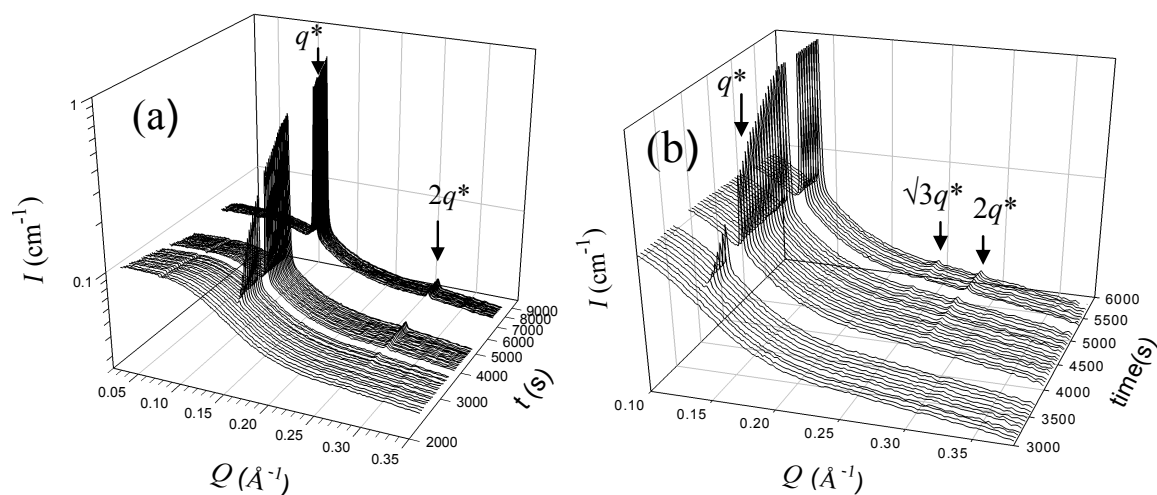


Figure S5: In-situ SAXS data measured for sealed solutions (of the reference sample composition) devoid of an air-water interface, displaying (a) L peaks at q^* and $2q^*$ at 25 °C and (b) H peaks at q^* , $\sqrt{3}q^*$, and $2q^*$ at 45 °C.