Electronic Supplementary Information for

Surfactant-mediated self-assembly of nanocrystals to form hierarchically structured zeolite thin films with controlled crystal orientation

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Fig. S1 Photographic image showing casting solutions with various concentration of TWEEN® 80.



Fig. S2 GIWAXS patterns of (a) bare silicon wafer substrate with water contact angle of 35° and (b) LTA-0.



Fig. S3 Top-view OM images (first row) and SEM images (second, third, and forth row) of LTA-12.5 (left column) and LTA-0 (right column) cast on a silicon wafer substrate with water contact angle of 35°.



Fig. S4 A statistical summary of the size of cylindrical patterns formed in the thin films. The thin films were cast on a silicon wafer substrate with (left column) a water contact angle of 54° and (right column) on the substrate with a water contact angle of 35° . The pattern size in the relatively hydrophobic silicon wafer (contact angle $=54^{\circ}$) is larger than that in the relatively hydrophilic silicon wafer (contact angle $=35^{\circ}$). It suggests that the size of the surfactant micelle can be strongly affected by the surface property of the substrate due to the difference in surface tension of the micelle.

LTA-12.5



Fig. S5 Top-view OM image (first column) and SEM image (second and third column) of zeolite LTA thin films cast on a glass substrate with water contact angle of 42°.



Fig. S6 Schematic illustration showing architecture of device used to measure electric properties of zeolite LTA thin films.



Fig. S7 Dielectric constants of (a) LTA-12.5 and (b) LTA-0 zeolite LTA thin films.



Fig. S8 Top-view OM images of zeolite LTA-9.67 on silicon wafer substrate with water contact angle of 35° (a) before and (b) after being treated by the high-pressure nitrogen.

Video S1

A video clip from the *in situ* investigation using an optical microscope is appended to gain insight into the formation of patterns in the zeolite LTA films via surfactant-mediated self-assembly (SMSA). The dot patterns in the spin cast zeolite solution were likely formed by micelles of the surfactant. The reversibility of the micelle formation during the temperature swing suggests that the surfactant micelle is very likely to be a thermodynamic phase.