## **Supplementary Information**

# Surface pressure induced 2D-crystallization of POM-based Surfactants : preparation of nanostructured thin films

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#### I. Compression-Expansion Cycles

As indicated in the figure below, the compression-expansion cycles curves beyond the crossover point all show strong hysteresis loop. However the hysteresis is fully reversible in the case of 2C12POM (Figure S1b) and C16POM, on the contrary to the others POMs surfactants (as C12POM, Figure S1b) for which a shift in the surface area is also observed.

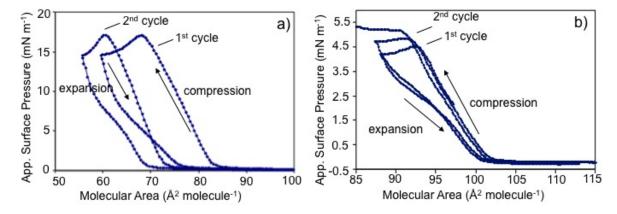


Figure S1: Compression-expansion cycles for (a) C12POM, and (b) 2C12POM, beyond the crossover point at room temperature and constant barrier speed (25 mm min<sup>-1</sup>).

#### II. C16 BAM images

Brewster Angle Microscopy has been used to assess the homogeneity of the interfacial layer at the micrometer scale for 2C12POM and C16POM Langmuir monolayer. As stated for 2C12POM, no features can be seen in the whole range of molecular area investigated for C16POM.

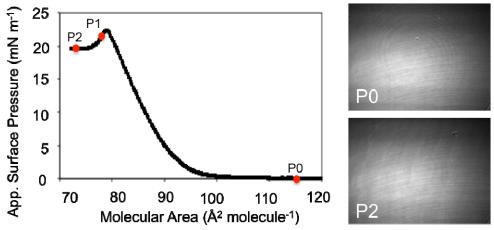


Figure S2: Apparent surface pressure versus molecular area isotherms for C16POM monolayers at the air/water interface at 20 °C (left) and corresponding BAM images (right).

### III. Layer transfer after one cycle

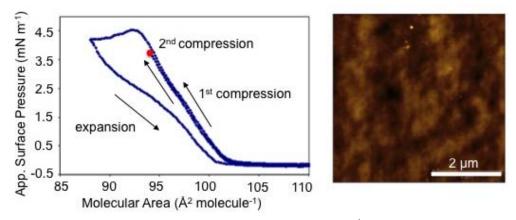


Figure S3: Transfer of a 2C12POM layer at 3.5 mN m<sup>-1</sup>, after one compression-expansion cycle. Apparent surface pressure versus molecular area isotherms (left) and corresponding AFM topography image (right).

IV Determination of the size repartition of the hexagonal crystallite structures from analysis of AFM images

The size repartition of the hexagonal crystallite structures was determined by the treatment of the AFM topography images of a 2C12POM layer transferred at 5 mN m<sup>-1</sup> after A<sub>c</sub>. (see Fig. 5 and Fig. S4). The image treatment was made a follows (1) splitting of the RGB color contribution, image treatment was made on the red contribution that gives the best contrast on the hexagonal structures, Fig. S4b (2) sharp points that do not participate to the hexagonal structures were suppressed using the shot-noise function (3) a filter with a threshold limit was applied to keep the darkest pixels, Fig. S4c (4) a Fourier transform process with a band pass filter between 9 and 40 pixels was applied to get rid of the large stripes present on the substrate, Fig. S4d (5) a second filter with a threshold limit was applied (6) a water shed treatment was applied in order to separate hexagons that appears nearly fused on the image (7) the plugin *analyze particle* was used to count and to obtain size of the hexagons (452), the hexagons being on the image border were not counted, Fig. S4e and Fig. 5.

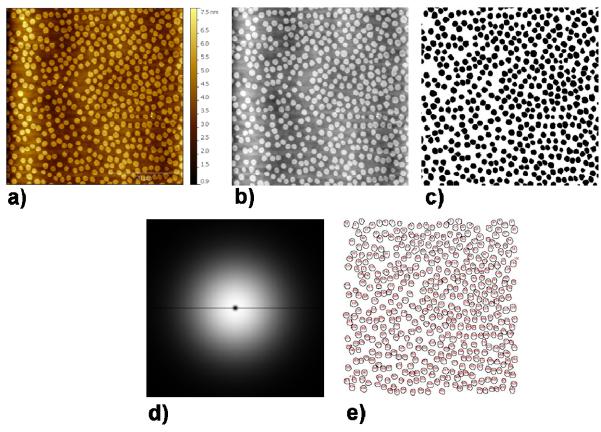


Figure S4: Image treatment of an AFM topography image of a 2C12POM layer showing hexagonal crystallite structures (see in the text for details).