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Supporting Information

for

Self-assembly, DNA complexation, and pH response of amphiphilic dendrimers
for gene transfection

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1. Complementary STM studies

Figure 12. STM images of **4** on HOPG. Dendrimer **4** was dissolved in CH₂Cl₂/MeOH 1:1 with a concentration of 0.4 mM, deposited by the solution-casting technique, and the sample annealed to 80°C. (A) Overview of a large scanned area (27 nm x 25 nm, -0.8 V, 1.2 pA). (B) Zoom (9.6 nm x 9.5 nm, -0.7 V, 1 pA).

Figure 13. Structure and STM image of *tert*-butylcarbamate (Boc) protected **8** on HOPG. The molecules were dissolved in CH₂Cl₂ (0.1 mM) and the sample was prepared by the solution-casting technique (30 nm x 30 nm, -0.7 V, 10 pA). Again, stripes with large and small periodicity can be

distinguished. Furthermore, the stripes are orthogonal to each other and the periodicities are the same within the error margins compared to **3** on HOPG.

Figure 14. Structure of the dimer **9** connecting two first-generation lipophilic dendrons and STM images of **9** on HOPG deposited by the solution-casting technique with a $\text{CH}_2\text{Cl}_2/\text{MeOH}$ 1:1 solution (0.4 mM). (a) Next to the ordered domain, a mobile phase is visible in the lower part of the image (14 nm x 15 nm, -1.1 V, 16 pA). (b) Higher resolution STM image displays the stripes of the small periodicity in detail (10 nm x 10 nm, -0.9 V, 28 pA).

2. BAM images

Figure 15. Representative BAM images of films on basic subphase.

Molecule **1** : (a) $A = 54 \text{ \AA}^2$, showing the coexistence of liquid domains (top) and aggregates (bottom). Contrast of the liquid domains has been digitally enhanced for better visibility. (b) $A = 18 \text{ \AA}^2$ and $\pi = 42.6 \text{ mN}\cdot\text{m}^{-1}$, (c) $A = 40 \text{ \AA}^2$, during decompression, showing the solid-like fracture of the film.

Molecule **2** : (d) $A = 55 \text{ \AA}^2$, showing the liquid state of the film. (e) $A = 30 \text{ \AA}^2$, $\pi = 30 \text{ mN}\cdot\text{m}^{-1}$.

Molecule **3** : (f) $A = 190 \text{ \AA}^2$, showing a liquid state with zones of unequal densities. (g) $A = 131 \text{ \AA}^2$ and $\pi = 18 \text{ mN}\cdot\text{m}^{-1}$.

Molecule **4** : (h) $A = 370 \text{ \AA}^2$, (i) $A = 118 \text{ \AA}^2$ and $\pi = 10 \text{ mN}\cdot\text{m}^{-1}$.

3. Langmuir Isotherms as a function of subphase pH

Figure 16. Isotherms of molecule **1**, recorded for different pH values of the subphase. Inset shows the reversibility of the isotherm for pH = 4.0.

Figure 17. Isotherms of molecule **3**, recorded for different pH values. Inset shows the reversibility of the isotherm for pH = 4.30 and pH = 11.5.

a)

b)

Figure 18. (a) Isotherms of molecule **4**, recorded for different pH values. Inset shows the reversibility of the isotherm for pH=11.5. (b) Sketch showing how a deeper immersion of the polar heads leads to an apparent decrease of the molecular area. The hydrophobic part of the molecules is not depicted, as it cannot be the limiting factor of compression. The maximum molecular area of the hydrophobic part of **4** is *ca.* 80 Å^2 whereas that of its polar head is *ca.* 110 Å^2 .

4. Langmuir Isotherms as a function of subphase DNA concentration

Figure 19. Isotherms of **1** on Tris-buffered water (pH 7.5, 20 °C), as a function of DNA concentration in the subphase.

Figure 20. Isotherms of **3** on Tris- buffered water (pH 7.5, 20 °C), as a function of DNA concentration in the subphase.

Figure 21. Isotherms of **4** on Tris- buffered water (pH 7.5, 20 °C), as a function of DNA concentration in the subphase.

5. "pH jump" experiment without DNA in the subphase

Figure 22. Surface pressure of **4** upon injection of HCl under a stabilized compressed film, for two values of the initial pressure (10 and 20 N/m).