## Supplementary Materials

## Surface Adsorption Properties and Layer Structures of Homogeneous Polyoxyethylene-Type Nonionic Surfactants in Quaternary-Ammonium-Salt-Type Amphiphilic Gemini Ionic Liquids with Oxygen- or Nitrogen-Containing Spacers

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## **Details of Measurements**

Wide-angle X-ray scattering (WAXS)



**Figure S1.** WAXS profiles for amphiphilic monomeric ionic liquids  $C_n$  NTf<sub>2</sub> at 25 °C: n = 4 (orange  $\bigcirc$ ), n = 6 (50 °C, yellow green  $\bigcirc$ ), n = 8 (blue  $\bigcirc$ ), n = 10 (purple  $\bigcirc$ ), and n = 12 (50 °C, red  $\bigcirc$ ).



**Figure S2.** Relationship between layer spacing  $d_1$  and alkyl chain length *n* for amphiphilic gemini and monomeric ionic liquids at 25 °C: 2C<sub>12</sub>(2-O-2) NTf<sub>2</sub> (black  $\blacksquare$ ) and C<sub>n</sub> NTf<sub>2</sub> (black  $\bigcirc$ , 50 °C for n = 6, 10).



**Figure S3.** Variation in (a)  $d_1$ , (b)  $d_2$ , and (c)  $d_3$  with temperature for amphiphilic ionic liquids: C<sub>12</sub> NTf<sub>2</sub> (black O), 2C<sub>12</sub>(2-O-2) NTf<sub>2</sub> (red  $\bullet$ ), 2C<sub>12</sub>(2-O-2-O-2) NTf<sub>2</sub> (orange  $\blacksquare$ ), 2C<sub>12</sub>(2-N-2) NTf<sub>2</sub> (blue  $\blacktriangle$ ), 2C<sub>12-3</sub> NTf<sub>2</sub> (green  $\blacklozenge$ ).



**Figure S4.** WAXS profiles for nonionic surfactant  $C_{12}EO_6$  in amphiphilic gemini ionic liquids at 25 °C: (a)  $2C_{12}(2\text{-}O-2\text{-}O-2)$  NTf<sub>2</sub>, (b)  $2C_{12}(2\text{-}N-2)$  NTf<sub>2</sub> and (c)  $2C_{12}(2\text{-}N-2)$  NTf<sub>2</sub> (30 °C): 0 (black  $\bigcirc$ ), 10 (gray  $\bigcirc$ ), 50 (blue  $\bigcirc$ ), 100 (yellow green  $\bigcirc$ ), 250 (yellow  $\bigcirc$ ), 500 (orange  $\bigcirc$ ), 750 (pink  $\bigcirc$ ), and 1000 mmol dm<sup>-3</sup> (red  $\bigcirc$ ).



**Figure S5.** Relationship between  $d_1$  and alkyl chain length of nonionic surfactants  $C_x EO_6$  at 1000 mmol dm<sup>-3</sup> in amphiphilic gemini ionic liquids:  $2C_{10}(2-O-2)$  NTf<sub>2</sub> (blue ),  $2C_{12}(2-O-2)$  NTf<sub>2</sub> (black ), and  $2C_{14}(2-O-2)$  NTf<sub>2</sub> (red ).



**Figure S6.** Relationship between  $d_1$  and alkyl chain length *n* of amphiphilic ionic liquid  $2C_n(2-O-2)$  NTf<sub>2</sub> containing nonionic surfactants  $C_x EO_6$  at 1000 mmol dm<sup>-3</sup>:  $C_8 EO_6$  (black  $\bullet$ ),  $C_{12} EO_6$  (red  $\blacksquare$ ), and  $C_{16} EO_6$  (blue  $\blacktriangle$ ).

Cryogenic transmission electron microscopy (cryo-TEM)



Figure S7. Cryo-TEM image of 750 mmol dm<sup>-3</sup>  $C_{12}EO_6$  in  $2C_{12}(2-O-2)$  NTf<sub>2</sub>.