

Electronic Supplementary Information for

## Regulating mesogenic properties of ionic liquid crystals by preparing binary or multi-component systems

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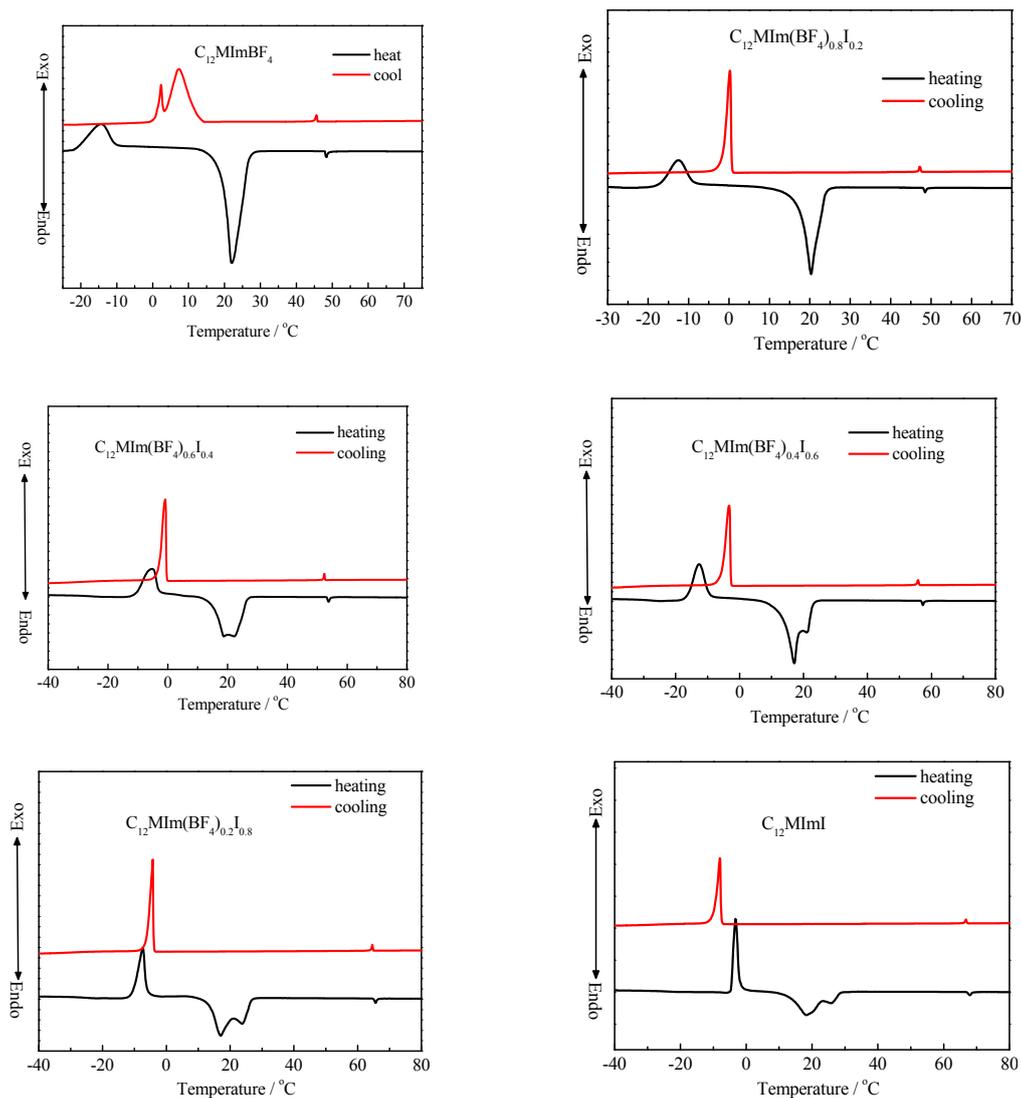
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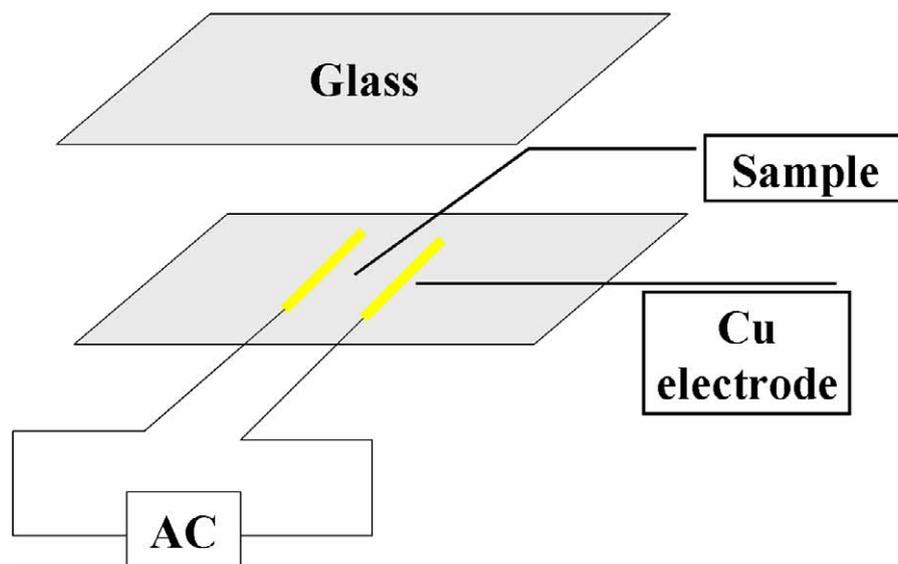
H-NMR of  $C_{12}MImBF_4$  and  $C_{12}MImI$

**$C_{12}MImI$ :**  $^1H$ -NMR(500M Hz,  $CD_3COCD_3$ ):  $\delta$  / ppm: 0.87 (3H, t,  $^3J = 6.5$  Hz,  $N(CH_2)_{11}-CH_3$ ), 1.27-1.37 (20H, m,  $N-CH_2-(CH_2)_{10}-CH_3$ ), 4.10 (3H, s,  $N-CH_3$ ), 4.42 (2H, t,  $^3J = 7.3$  Hz,  $N-CH_2-C_{11}H_{23}$ ), 7.78 (1H, s,  $-N(C_{12}H_{25})-CH=$ ), 7.85 (1H, s,  $-N(C_{12}H_{25})-CH=N(CH_3)-$ ), 9.53 (1H, s,  $-N(CH_3)-CH=N(C_{12}H_{25})$ ).

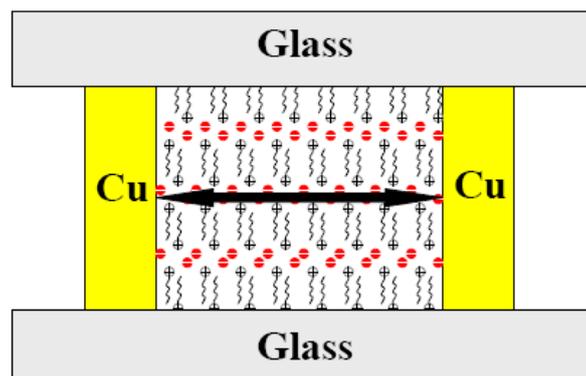
**$C_{12}MImBF_4$ :**  $^1H$ -NMR(500M Hz,  $CD_3COCD_3$ ):  $\delta$  / ppm: 0.92 (3H, t,  $J = 7.5$  Hz,  $N(CH_2)_{11}-CH_3$ ), 1.27-1.47 (18H, m,  $N-CH_2-CH_2-(CH_2)_9-CH_3$ ), 1.69 (2H, m,  $-NCH_2-CH_2-(CH_2)_9-CH_3$ ), 4.18 (3H, s,  $-N-CH_3$ ), 4.40 (2H, t,  $^3J = 6.0$  Hz,  $-N-CH_2-C_{11}H_{23}$ ), 7.66 (1H, d,  $^3J = 3.0$  Hz,  $-N(C_{12}H_{25})-CH=$ ), 7.75 (1H, d,  $^3J = 3.0$  Hz,  $-N(C_{12}H_{25})-CH=N(CH_3)-$ ), 9.10 (1H, s,  $-N(CH_3)-CH=N(C_{12}H_{25})$ ).



**Fig. S1** DSC heating and cooling traces of the binary system  $C_{12}MIm(BF_4)_yI_x$ .



**Fig. S2** Schematic drawing of the cell using Cu electrodes for the measurement of the conductivity parallel to the smectic layer ( $\sigma_{\parallel}$ ).



**Fig. S3** Schematic drawing of the alignment of ILC in the cell using Cu electrodes.

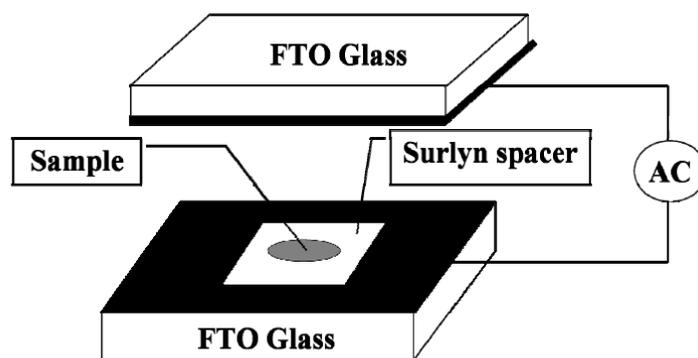


Fig. S4 Schematic drawing of the cell using FTO electrodes for the measurement of the conductivity perpendicular to the smectic layer ( $\sigma_{\perp}$ ).

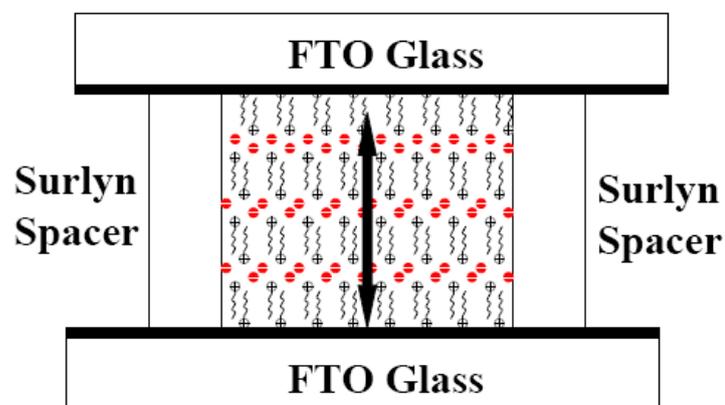
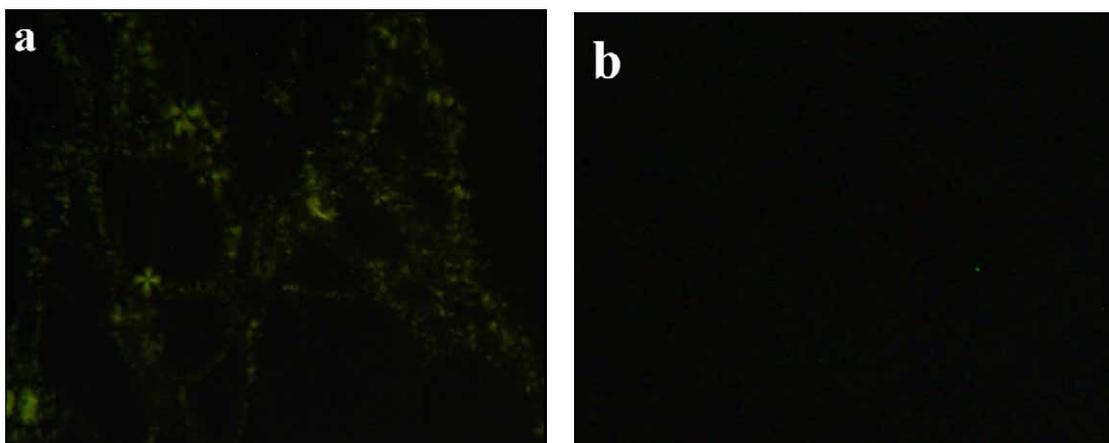
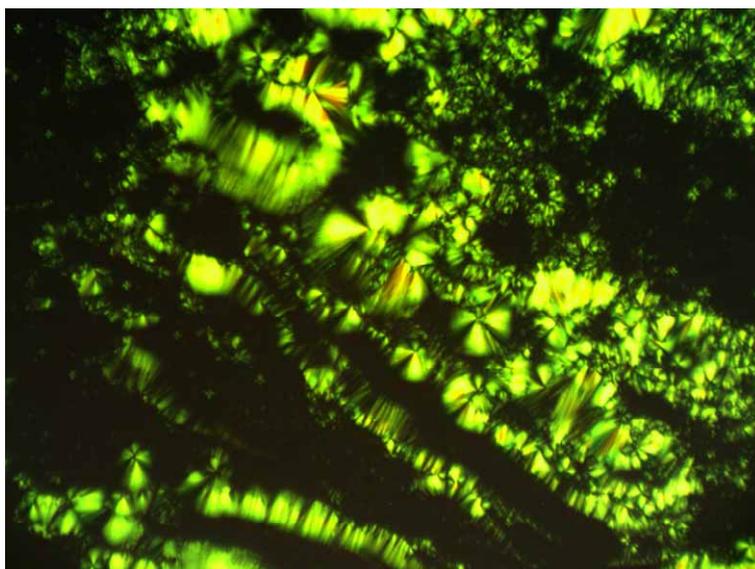


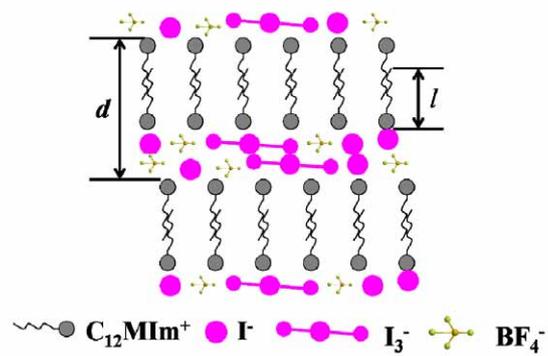
Fig. S5 Schematic drawing of the alignment of ILCs in the cell using FTO electrodes.



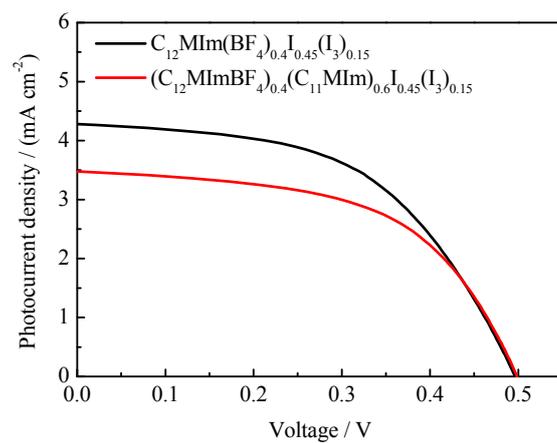
**Fig. S6** Polarized optical microscopic textures of  $C_{12}MIm(BF_4)_{0.4}I_{0.6}$  observed at 35 °C between two pieces of glass substrate for conductivity measurement (a) before and (b) after a pressure was applied.



**Fig. S7** Polarized optical microscopic texture of  $C_{12}MIm(BF_4)_{0.4}I_{0.45}(I_3)_{0.15}$  at 300 K.



**Fig. S8** Schematic illustration of the interdigitated bilayer structure of  $C_{12}MIm(BF_4)_{0.4}I_{0.45}(I_3)_{0.15}$  in SmA liquid crystalline phase.



**Fig. S9** *J-V* curves of DSCs with different electrolytes.