Electronic Supplementary information

The Roles of Betaine - Ester Analogues of 1-*N*-alkyl-3-*N*'-methyl Imidazolium Salts: As Amphotropic Ionic Liquid Crystals, and Organogelators

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Experimental Section.

Instrumentation. The ^{1}NMR and ^{13}CNMR spectra were recorded on Bruker Advance DXP$_{400}$ spectrometer in $CDCl_3$. The solvents used were reagent grade and without further purification. IR spectra were recorded with a Perkin-Elmer Spectrum One. Optical characterization was performed by using covered microscope slides on an Olympus BH-2 polarizing microscope equipped with a Mettler Toledo FP82 hot stage and a Mettler Toledo FP90 central processor. Phase transition temperatures were determined by differential scanning calorimetry (DSC) at a scan rate of 10 ^{o}C/min using Mettler Toledo DSC822 calorimeter calibrated under flowing N_2 with indium and tin standards. The decomposition temperatures were determined by thermo gravimetric analysis using a Mettler Toledo TGA851. The powder X-ray diffraction patterns were obtained by National Dong Hwa University with Cu Kαradiation (XRD D8Advanced, Bruker) and by National Synchrotron Radiation Research Center. Scanning electron microscopy (SEM) images were obtained using a JEOL JEM-6500F operated at 15 KV. The UV-Visible absorption spectra of the Ag-NPs dispersions were obtained using PerkinElmer Lambda 750 spectrophotometer. DLS and ZP measurements were performed on different concentrations of lyotropic LC solutions at room temperature using a Zetasizer Nano ZS ZEN 3600 instrument. The images of nanoparticles were acquired by an Analytical Transmission Electron Microscope (JEOL JEM-3010).

Octyl 2-bromoacetate: Colorless liquid (Yield: 79%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 4.12 (t, J = 8 Hz, 2H, CH_2), 3.79 (s, 2H, CH_2), 1.61 (t, J = 8 Hz, 2H, CH_2), 1.23 (m, 10H, CH_2), 0.83 (t, J = 8 Hz, 3H, CH_3).

Decyl 2-bromoacetate: Colorless liquid (Yield: 76%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 4.12 (t, J = 8 Hz, 2H, CH_2), 3.79 (s, 2H, CH_2), 1.62 (J = 8 Hz, 2H, CH_2), 1.22 (m, 14H, CH_2), 0.84 (t, J = 8 Hz, 3H, CH_3).

Dodecyl 2-bromoacetate: Colorless liquid (Yield: 79%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.38 (t, J = 8 Hz, 2H, CH_2), 1.84 (t, J = 8 Hz, 2H, CH_2), 1.25 (m, 18H, CH_2), 0.87 (t, J = 8 Hz, 3H, CH_3).

Tetradecyl 2-bromoacetate: Colorless liquid (Yield: 81%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 4.14 (t, J = 8 Hz, 2H, CH_2), 3.80 (s, 2H, CH_2), 1.63 (J = 8 Hz, 2H, CH_2), 1.23 (m, 22H, CH_2), 0.85 (t, J = 8 Hz, 3H, CH_3).

Hexadecyl 2-bromoacetate: Colorless liquid (Yield: 83%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 4.16 (t, J = 8 Hz, 2H, CH_2), 3.83 (s, 2H, CH_2), 1.65 (J = 8 Hz, 2H, CH_2), 1.25 (m, 26H, CH_2), 0.87 (t, J = 8 Hz, 3H, CH_3).

Octadecyl 2-bromoacetate: Colorless liquid (Yield: 88%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 4.16 (t, J = 8 Hz, 2H, CH_2), 3.83 (s, 2H, CH_2), 1.65 (J = 8 Hz, 2H, CH_2), 1.25 (m, 30H, CH_2), 0.87 (t, J = 8 Hz, 3H, CH_3).

1-methyl-3-octaneoxyoxoethyl-imidazolium bromide [C_1, EC_8-Im]Br · 2.5 H_2O. Sticky solid. (Yield: 2.54g, 77%).^{1}H NMR (400 MHz, $CDCl_3$): δ 9.54 (s, 1H, Ar-H), 7.55 (s, 2H, Ar-H), 5.35 (s, 2H, CH_2), 4.14 (t, J = 8 Hz, 2H, CH_2), 4.00 (s, 3H, CH_3), 1.62 (t, J = 8 Hz, 2H, CH_2), 1.24 (m, 10H, CH_2), 0.84 (t, J = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.61 (C=O), 137.94 (NCN), 123.67 (NCHCHN), 123.42 (NCHCHN), 66.97 (NCH_3), 50.34 (NCH_2), 36.97 (CH_2), 31.75 (CH_2), 29.15 (CH_2), 29.13 (CH_2), 28.34 (CH_2), 25.69 (CH_2), 22.60 (CH_2), 14.05 (CH_3). Elemental analysis: Anal. Calcd for $C_{14}H_{30}BrN_2O_{4.5}$: C, 44.45; H, 7.99; N, 7.41. Found: C, 44.30; H, 8.02; N, 7.41.

1-methyl-3-decaneoxyoxoethyl-imidazolium bromide [C_1, EC_{10}-Im]Br · 1H_2O. Sticky solid. (Yield: 2.73g, 77%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.41 (s, 1H, Ar-H), 7.57 (s, 1H, Ar-H), 7.54 (s, 1H, Ar-H), 5.32 (s, 2H, CH_2), 4.13 (t, J = 4Hz, 2H, CH_2), 3.97 (s, 3H, CH_3), 1.61 (t, J = 8 Hz, 2H, CH_2), 1.22 (m, 14H, CH_2), 0.83 (t, J = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.76 (C=O), 137.78 (NCN), 123.65 (NCHCHN), 123.54 (NCHCHN), 66.93 (NCH_3), 50.31 (NCH_2), 36.93 (CH_2), 31.86 (CH_2), 29.55 (CH_2), 29.52 (CH_2), 29.29 (CH_2), 29.25 (CH_2), 28.36 (CH_2), 25.70 (CH_2), 22.64 (CH_2), 14.07 (CH_3). Elemental analysis : Anal. Calcd for $C_{16}H_{31}BrN_2O_3$: C, 50.66; H, 8.24; N, 7.38. Found: C, 50.63; H, 8.09; N 7.40.

1-methyl-3-dodecaneoxyoxoethyl-imidazolium bromide [C_1, EC_{12}-Im]Br · 1H_2O. Sticky solid. (Yield: 2.73g, 74%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 10.01 (s, 1H, Ar-H), 7.53 (s, 1H, Ar-H), 7.44 (s, 1H, Ar-H), 5.43 (s, 2H, CH_2), 4.19 (t, J = 8 Hz, 2H, CH_2), 4.08 (s, 3H, CH_3), 1.67 (t, J = 8 Hz, 2H, CH_2), 1.26 (m, 18H, CH_2), 0.88 (t, J = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.23 (C=O), 138.57 (NCN), 123.61 (NCHCHN), 122.91 (NCHCHN), 67.11 (NCH_3), 50.34 (NCH_2), 36.93 (CH_2), 31.89 (CH_2), 29.62 (CH_2), 29.57 (CH_2), 29.47 (CH_2), 29.33 (CH_2), 29.18 (CH_2), 28.35 (CH_2), 25.70 (CH_2), 22.67 (CH_2), 14.10 (CH_3). Elemental analysis: Anal. Calcd for $C_{18}H_{35}BrN_2O_3$: C, 53.07; H, 8.66; N, 6.88. Found: C, 53.46; H, 8.54; N 6.84.

1-methyl-3-tetradecaneoxyoxoethyl-imidazolium bromide [C_1, EC_{14}-Im]Br · 1H_2O. White Solid (Yield: 2.03g, 79%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.99 (s, 1H, Ar-H), 7.59 (s, 1H, Ar-H), 7.51 (s, 1H, Ar-H), 5.41 (s, 2H, CH_2), 4.14 (t, J = 8 Hz, 2H, CH_2), 4.05 (s, 3H, CH_2), 1.62 (t, J = 8 Hz, 2H, CH_2), 1.21 (m, 22H, CH_2), 0.83 (t, J = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ

166.17 (C=O), 138.59 (NCN), 123.64 (NCHCHN), 122.89 (NCHCHN), 67.13 (NCH_3), 50.43 (NCH_2), 36.93 (CH_2), 31.90 (CH_2), 29.66 (CH_2), 29.64 (CH_2), 29.58 (CH_2), 29.48 (CH_2), 29.34 (CH_2), 29.18 (CH_2), 28.35 (CH_2), 25.71 (CH_2), 22.67 (CH_2), 14.10 (CH_3). Elemental analysis: Anal. Calcd for $C_{20}H_{39}BrN_2O_3$: C, 55.17; H, 9.03; N, 6.43. Found: C, 55.54; H, 9.21; N, 6.25.

1-methyl-3-hexadecaneoxyoxoethyl-imidazolium bromide [C_1, EC_{16}-Im]Br · $1H_2O$. White Solid (Yield: 2.35g, 78%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.87 (s, 1H, Ar-H), 7.58 (s, 1H, Ar-H), 7.52 (s, 1H, Ar-H), 5.40 (s, 2H, CH_2), 4.14 (t, *J* = 8 Hz, 2H, CH_2), 4.04 (s, 3H, CH_2), 1.62 (t, *J* = 8 Hz, 2H, CH_2), 1.21 (m, 26H, CH_2), 0.83 (t, *J* = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.15 (C=O), 137.91 (NCN), 123.83 (NCHCHN), 123.25 (NCHCHN), 66.90 (NCH_3), 50.17 (NCH_2), 36.83 (CH_2), 31.80 (CH_2), 29.59 (CH_2), 29.40 (CH_2), 29.24 (CH_2), 29.12 (CH_2), 28.24 (CH_2), 25.61 (CH_2), 22.56 (CH_2), 14.02 (CH_3). Elemental analysis: Anal. Calcd for $C_{22}H_{43}BrN_2O_3$: C, 57.01; H, 9.35; N, 6.04. Found: C, 57.27; H, 9.35; N, 6.28.

1-methyl-3-octadecaneoxyoxoethyl-imidazolium bromide [C_1, EC_{18}-Im]Br · $1H_2O$. White Solid (Yield: 2.58g, 81%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.96 (s, 1H, Ar-H), 7.59 (s, 1H, Ar-H), 7.52 (s, 1H, Ar-H), 5.41 (s, 2H, CH2), 4.14 (t, *J* = 8 Hz, 2H, CH_2), 4.05 (s, 3H, CH_2), 1.62 (t, *J* = 8 Hz, 2H, CH_2), 1.21 (m, 30H, CH_2), 0.83 (t, *J* = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.20 (C=O), 138.29 (NCN), 123.76 (NCHCHN), 123.09 (NCHCHN), 67.05 (NCH_3), 50.31 (NCH_2), 36.92 (CH_2), 31.89 (CH_2), 29.67 (CH_2), 29.48 (CH_2), 29.32 (CH_2), 29.19 (CH_2), 28.34 (CH_2), 25.69 (CH_2), 22.65 (CH_2), 14.08 (CH_3). Elemental analysis : Anal. Calcd for $C_{24}H_{47}BrN_2O_3$: C, 58.64; H, 9.64; N, 5.70. Found: C, 58.66; H, 9.65; N, 5.67.

Synthesis of 1-methyl-3-dodecaneoxyoxoethyl-imidazolium BF_4 [C_1, EC_{12}-Im]BF_4. A flask was charged with [C_1,EC_{12}-Im]Br (0.5g, 1.28 mmol) and 10 mL MeOH. NH_4BF_4 (0.135 g, 1.28 mmol) was dissolved in 30 mL of MeOH in the other flask and dropping into [C_1,EC_{12}-Im]Br solution. After 2 hours the precipitate was filtered and recrystallized from DCM/Ether to afford the white crystalline compounds (Yield: 0.47 g, 92%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.64 (s, 1H, Ar-H), 7.36 (s, 1H, Ar-H), 7.34 (s, 1H, Ar-H), 4.99 (s, 2H, CH_2), 4.15 (t, *J* = 8 Hz, 2H, CH_2), 3.87 (s, 3H, CH_3), 1.64 (t, *J* = 8 Hz, 2H, CH_2) , 1.25 (m, 18H, CH_2), 0.86 (t, *J* = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.24 (C=O), 137.64 (NCN), 123.67 (NCHCHN), 123.30 (NCHCHN), 66.99 (NCH_3), 49.77 (NCH_2), 36.30 (CH_2), 31.90 (CH_2), 29.63 (CH_2), 29.51 (CH_2), 29.34 (CH_2), 29.23 (CH_2), 28.32 (CH_2), 25.68 (CH_2), 22.67 (CH_2), 14.09 (CH_3). Elemental analysis: Anal. Calcd for $C_{18}H_{33}BF_4N_2O_2$: C, 54.56; H, 8.39; N, 7.07. Found: C, 54.64; H, 8.49; N, 7.05.

The following compounds were prepared in the same way.

1-methyl-3-tetradecaneoxyoxoethyl-imidazolium BF_4 [C_1, EC_{14}-Im]BF_4. White solid (Yield: 0.45g, 90%). ^{1}H NMR (400MHz, $CDCl_3$): δ 8.72 (s, 1H, Ar-H), 7.35 (s, 1H, Ar-H), 7.32 (s, 1H, Ar-H), 5.01 (s, 2H, CH_2), 4.18 (t, *J* = 4Hz, 2H, CH_2), 3.91 (s, 3H, CH_3), 1.67 (t, *J* = 8 Hz, 2H, CH_2) , 1.25 (m, 22H, CH_2), 0.87 (t, *J* = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.16 (C=O), 137.78 (NCN), 123.64 (NCHCHN), 123.18 (NCHCHN), 67.06 (NCH_3), 49.82 (NCH_2), 36.38 (CH_2), 31.92 (CH_2), 29.69 (CH_2), 29.66 (CH_2), 29.62 (CH_2), 29.51 (CH_2), 29.36 (CH_2), 29.22 (CH_2), 28.32 (CH_2), 25.69 (CH_2), 22.68 (CH_2), 14.10 (CH_3). Elemental analysis: Anal. Calcd for $C_{20}H_{37}BF_4N_2O_2$: C, 56.61; H, 8.79; N, 6.60. Found: C, 56.27; H, 8.76; N, 6.55.

1-methyl-3-hexadecaneoxyoxoethyl-imidazolium BF_4 [C_1, EC_{16}-Im]BF_4. White solid (Yield: 0.47g, 93%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.44 (s, 1H, Ar-H), 7.30 (s, 1H, Ar-H), 7.28 (s, 1H, Ar-H), 4.92 (s, 2H, CH_2), 4.17 (t, J = 8 Hz, 4H, CH_2), 3.85 (s, 3H, CH_3), 1.65 (t, J = 8 Hz, 2H, CH_2), 1.25 (m, 26H, CH_2), 0.87 (t, J = 8 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.26 (C=O), 137.67 (NCN), 123.66 (NCHCHN), 123.25 (NCHCHN), 67.03 (NCH_3), 49.77 (NCH_2), 36.34 (CH_2), 31.94 (CH_2), 29.74 (CH_2), 29.68 (CH_2), 29.56 (CH_2), 29.44 (CH_2), 29.39 (CH_2), 29.26 (CH_2), 28.32 (CH_2), 25.70 (CH_2), 22.71 (CH_2), 14.14 (CH_3). Elemental analysis: Anal. Calcd for $C_{22}H_{41}BF_4N_2O_2$: C, 58.41; H, 9.14; N, 6.19. Found: C, 58.62; H, 9.08; N, 6.16.

Synthesis of 1-methyl-3-dodecaneoxyoxoethyl-imidazolium PF_6 [C_1, EC_{12}-Im]PF_6. A flask was charged with [C_1, EC_{12}-im]Br (0.5g, 1.10 mmol) and 10 mL MeOH. NH_4PF_6 (0.18g, 1.10 mmol) was dissolved in 30 mL of MeOH in the other flask and dropping into [C_1,EC_{12}-im]Br solution. After 2 hours, the precipitate was filtered and recrystallized from DCM/Ether to afford the white crystalline compounds (Yield: 0.54g, 93%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.55 (s, 1H, Ar-H), 7.31 (s, 1H, Ar-H), 7.27 (s, 1H, Ar-H), 4.95 (s, 2H, CH_2), 4.20 (t, J = 8Hz, 2H, CH_2), 3.91 (s, 3H, CH_3), 1.66 (t, J = 8 Hz, 2H, CH_2), 1.26 (m, 18H, CH_2), 0.87 (t, J = 8 Hz, 6H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.24 (C=O), 137.64 (NCN), 123.67 (NCHCHN), 123.30 (NCHCHN), 66.99 (NCH_3), 49.77 (NCH_2), 36.30 (CH_2), 31.90 (CH_2), 29.63 (CH_2), 29.51 (CH_2), 29.34 (CH_2), 29.23 (CH_2), 28.32 (CH_2), 25.68 (CH_2), 22.67 (CH_2), 14.09 (CH_3). Elemental analysis: Anal. Calcd for $C_{18}H_{33}F_6N_2O_2P$: C, 47.57; H, 7.32; N, 6.16. Found: C, 47.58; H ,7.37; N, 6.17.

The following compounds were prepared in the same way.

1-methyl-3-tetradecaneoxyoxoethyl-imidazolium PF_6 [C_1, EC_{14}-Im]PF_6. White solid (Yield: 0.53g, 93%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.58 (s, 1H, Ar-H), 7.31 (s, 1H, Ar-H), 7.22 (s, 1H, Ar-H), 4.96 (s, 2H, CH_2), 4.21 (t, J = 8 Hz, 2H, CH_2), 3.94 (s, 3H, CH_3),1.67 (t, J = 8 Hz, 2H, CH_2), 1.25 (m, 22H, CH_2), 0.87 (t, J = 8 Hz, 6H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 165.98 (C=O), 137.24 (NCN), 123.60 (NCHCHN), 123.29 (NCHCHN), 67.11 (NCH_3), 49.73 (NCH_2), 36.27 (CH_2), 31.92 (CH_2), 29.71 (CH_2), 29.52 (CH_2), 29.36 (CH_2), 29.23 (CH_2), 29.28 (CH_2), 25.67 (CH_2), 22.68 (CH_2), 14.10 (CH_3). Elemental analysis: Anal. Calcd for $C_{20}H_{37}F_6N_2O_2P$: C, 49.79; H, 7.73; N, 5.81. Found: C, 49.52; H, 8.01; N, 5.72.

1-methyl-3-hexadecaneoxyoxoethyl-imidazolium PF_6 [C_1, EC_{16}-Im]PF_6. White solid (Yield: 0.55 g, 96%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.54 (s, 1H, Ar-H), 7.31 (s, 1H, Ar-H), 7.27 (s, 1H, Ar-H), 4.95 (s, 2H, CH_2), 4.20 (t, J = 8 Hz, 2H, CH_2), 3.92 (s, 3H, CH_2), 1.66 (t, J = 8 Hz, 2H, CH_2), 1.25 (m, 26H, CH_2), 0.87 (t, J = 4 Hz, 3H, CH_3). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.26 (C=O), 137.66 (NCN), 123.66 (NCHCHN), 123.25 (NCHCHN), 67.02 (NCH_3), 49.77 (NCH_2), 36.34 (CH_2), 31.93 (CH_2), 29.73 (CH_2), 29.68 (CH_2), 29.55 (CH_2), 29.38 (CH_2), 29.26 (CH_2), 28.32 (CH_2), 25.70 (CH_2), 22.70 (CH_2), 14.14 (CH_3). Elemental analysis: Anal. Calcd for $C_{22}H_{41}F_6N_2O_2P$: C, 51.76; H, 8.09; N, 5.49. Found: C, 51.85; H, 8.00; N, 5.43.

Preparation of Ag-NPs. A lyotropic LC solution sample was prepared with 40 wt% [C_1, EC_{16}-Im] Br (0.21 g, 0.47 mmol) in $CHCl_3$. Similarly another LC solution sample has been prepared and then heated to 60 ^{o}C and cooled back to RT to prepare LC gel sample. $AgNO_3$ (0.008 g, 0.047 mmol) and subsequently 2 drops of MeOH were added to both samples. The reaction mixtures were then kept under stirring, and 30 mL of DI water was added in each and then

dropping the 5mL of an aqueous $NaBH_4$ (0.018g, 0.47 mmol) into solutions. Once all reagents were combined, both the sample vials were allowed for stirring 10 min to fully mix the reactants. Then, immediately the colourless mixture turns to deep yellow for LC solution sample, while the change from colourless to yellow to deep brown was observed in the LC gel sample, indicating the formation of Ag-NPs. The dispersed Ag-NPs in all solutions were extracted into $CHCl_3$ layer by adding each 30 mL of DI water and chloroform. Then, the resulting NPs were collected by centrifugation, washed three times with methanol and suspended in $CHCl_3$. Each sample was transferred into two vials with one stored in dark and another under light for comparison.

Table S1. Crystal data and structure refinement for [C_1, EC_{16}-Im] Br

Identification code	1	
Empirical formula	C_{22} H_{41} Br N_2 O_2	
Formula weight *(M)*	445.48	
Temperature	296(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	P2(1)/c	
Unit cell dimensions	a = 22.834(7)Å	α=90 deg
	b = 8.194(3)Å	β=98.048(8) deg
	c = 13.429(4)Å	γ=90 deg
Volume	2488.0(13)A^3	
Z	4	
Density (calculated)	1.189 mg/m^3	
Absorption coefficient	1.669 mm^{-1}	
F (000)	952	
Crystal size	0.23 x 0.21 x 0.09 mm	
Theta range for data collection	0.90 to 25.31 deg.	
Index ranges	-27<=h<=26, -9<=k<=9, -16<=l<=13	
Reflections collected	11675 / 4450 [R(int) = 0.0795]	
Completeness to theta =	98.2 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.8643 and 0.7001	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	4450 / 0 / 246	
Goodness-of-fit on F^2	0.969	
Final R indices [I > 2 *sigma (I)*]	R1 = 0.0478, wR_2 = 0.1059	
R indices (all data)	R1 = 0.1513, wR_2 = 0.1607	
Largest diff. peak and hole	0.417 and -0.590 $e.A^{-3}$	

Table S2. Crystal data and structure refinement for [C_1, EC_{16}-Im] BF_4

Identification code	2	
Empirical formula	C_{22} H_{41} N_2 O_2 B F_4	
Formula weight *(M)*	452.38	
Temperature	296(2) K	
Wavelength	0.71073 A	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 8.4947(7) Å	α= 82.735(4) deg
	b = 9.0566(7) Å	β= 88.304(4) deg
	c = 32.905(3) Å	γ= 88.047(4) deg
Volume	2508.9(4) A^3	
Z	4	
Density (calculated)	1.201 Mg/m^3	
Absorption coefficient	0.099 mm^{-1}	
F (000)	976	
Crystal size	0.70 x 0.25 x 0.25 mm	
Theta range for data collection	1.87 to 25.00 deg.	
Index ranges	-10<=h<=10, -11<=k<=11, -40<=l<=41	
Reflections collected	32516 / 8822 [R(int) = 0.0569]	
Completeness to theta =	99.8 %	
Absorption correction	None	
Max. and min. transmission	0.9671 and 0.9118	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	8822 / 0 / 571	
Goodness-of-fit on F^2	1.055	
Final R indices [I > *2* sigma (I)]	R1 = 0.0748, wR_2 = 0.1705	
R indices (all data)	R1 = 0.1433, wR_2 = 0.1694	
Largest diff. peak and hole	0.625 and -0.511 $e.A^{-3}$	

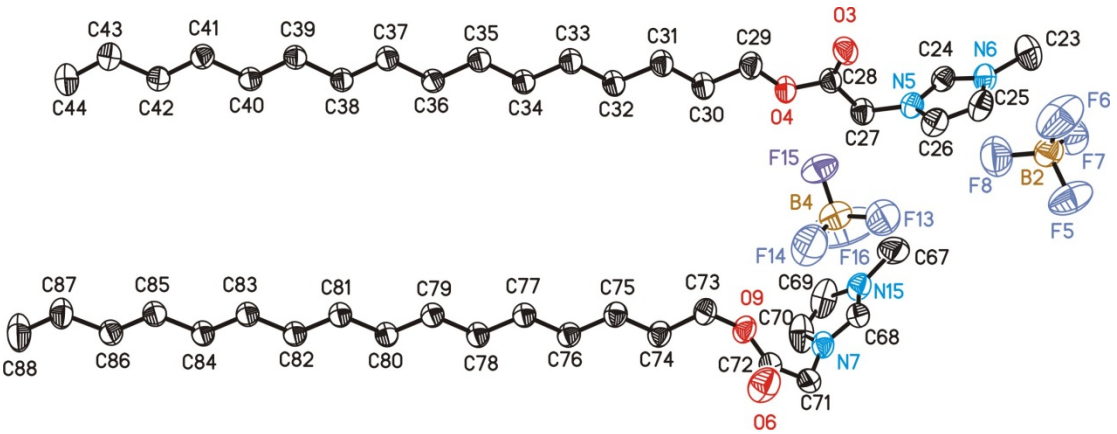

Figure S1. The ORTEP drawing structure of $[C_1, EC_{16}\text{-Im}]BF_4$ (50% thermal ellipsoids), hydrogens being omitted for clarity.

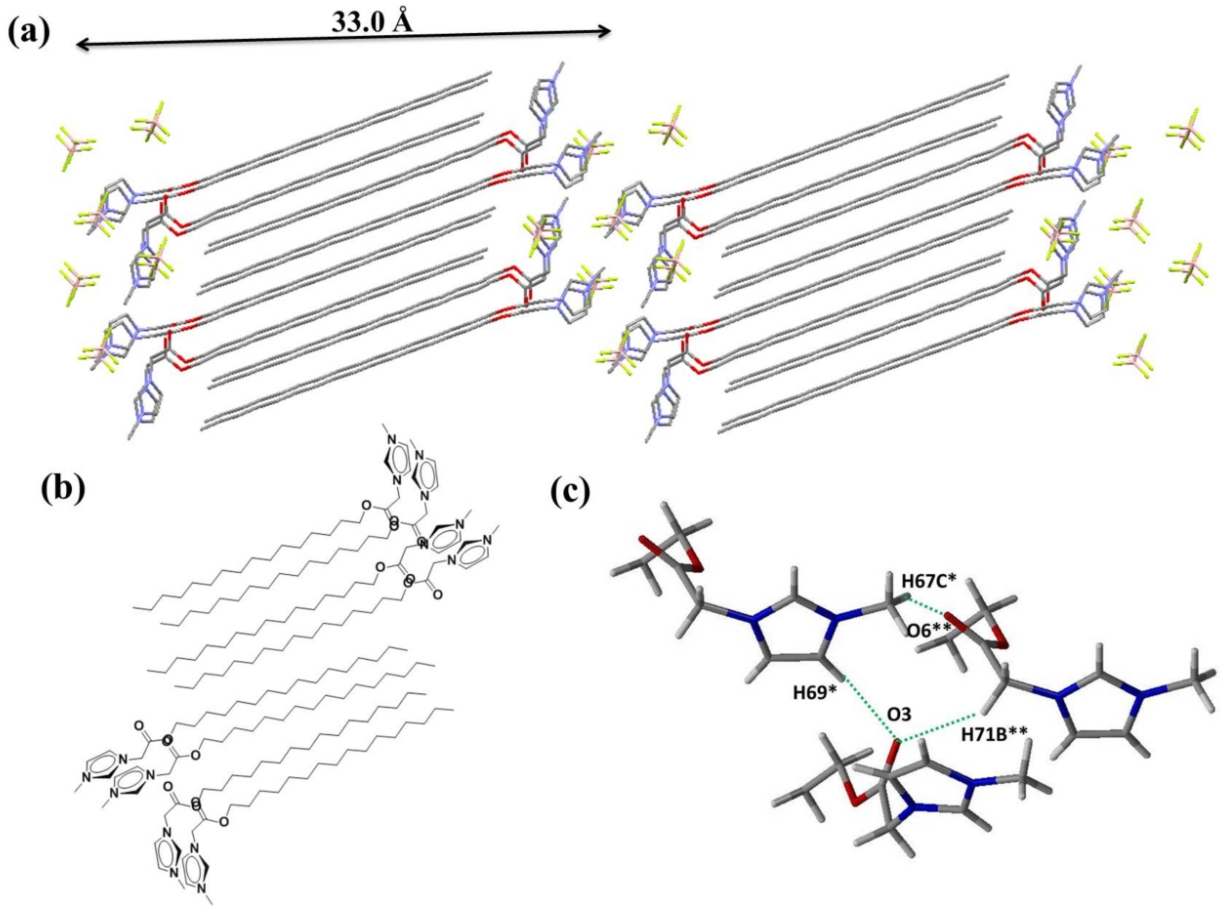

Figure S2. (a) Crystal packing of $[C_1, EC_{16}\text{-Im}]BF_4$, (b) structure packing profile of $[C_1, EC_{16}\text{-Im}]BF_4$; (c) the close interactions around two different C=O groups have dimensions (H69*…O3: 2.64 Å; O3...H71B** : 2.70 Å; H67C* ···O6** : 2.47 Å) where * is symmetry operation (x, -1+y z) and ** is symmetry operation (1+x, -1+y z).

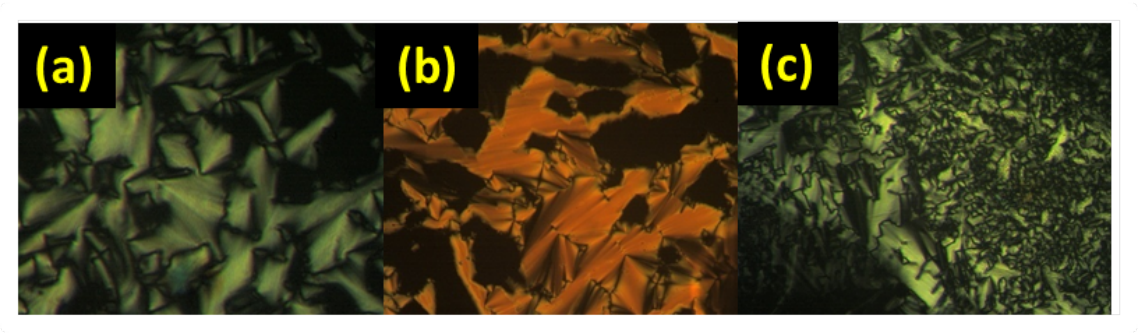

Figure S3. POM images of (a) [C_1, EC_{16}-Im]Br, (b) [C_1, EC_{16}-Im]BF_4, (c) [C_1, EC_{16}-Im]PF_6 in the first cooling process at 120℃.

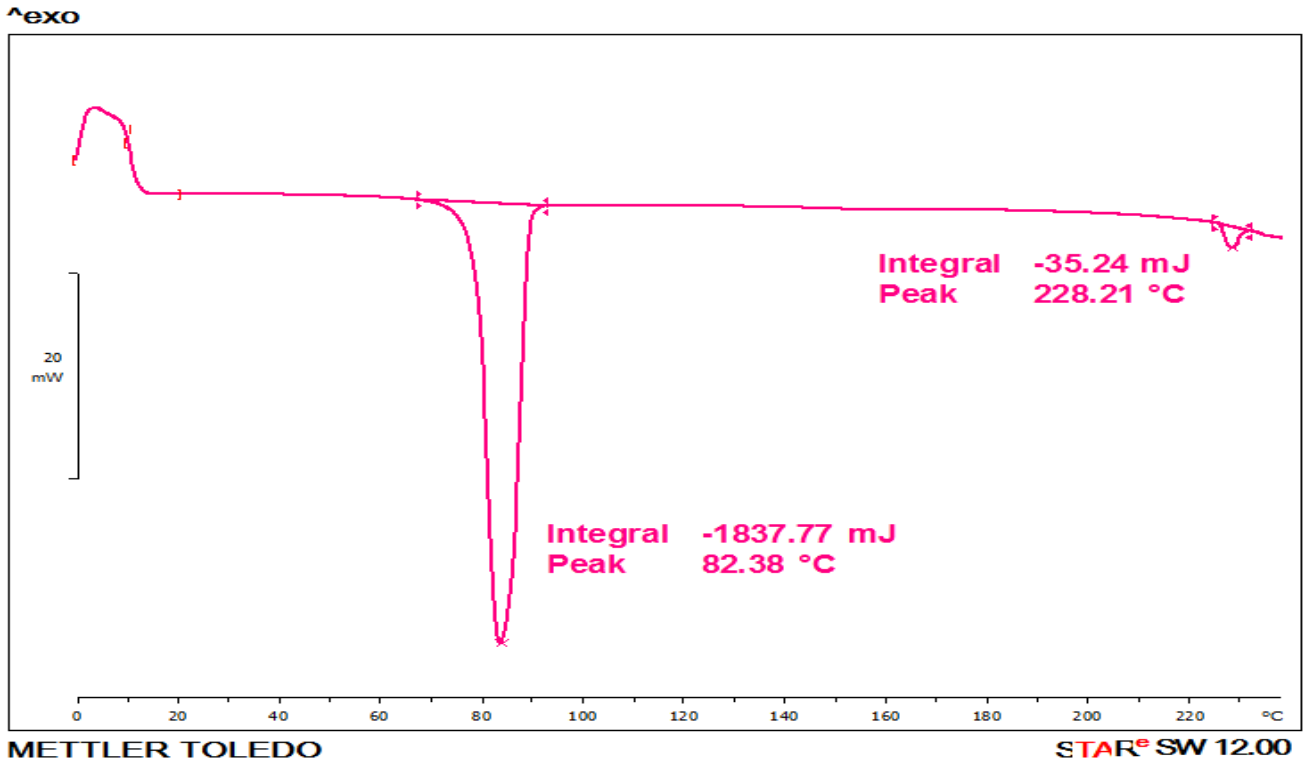

Figure S4 (a). DSC thermogram for [C_1, EC_{14}-Im]Br

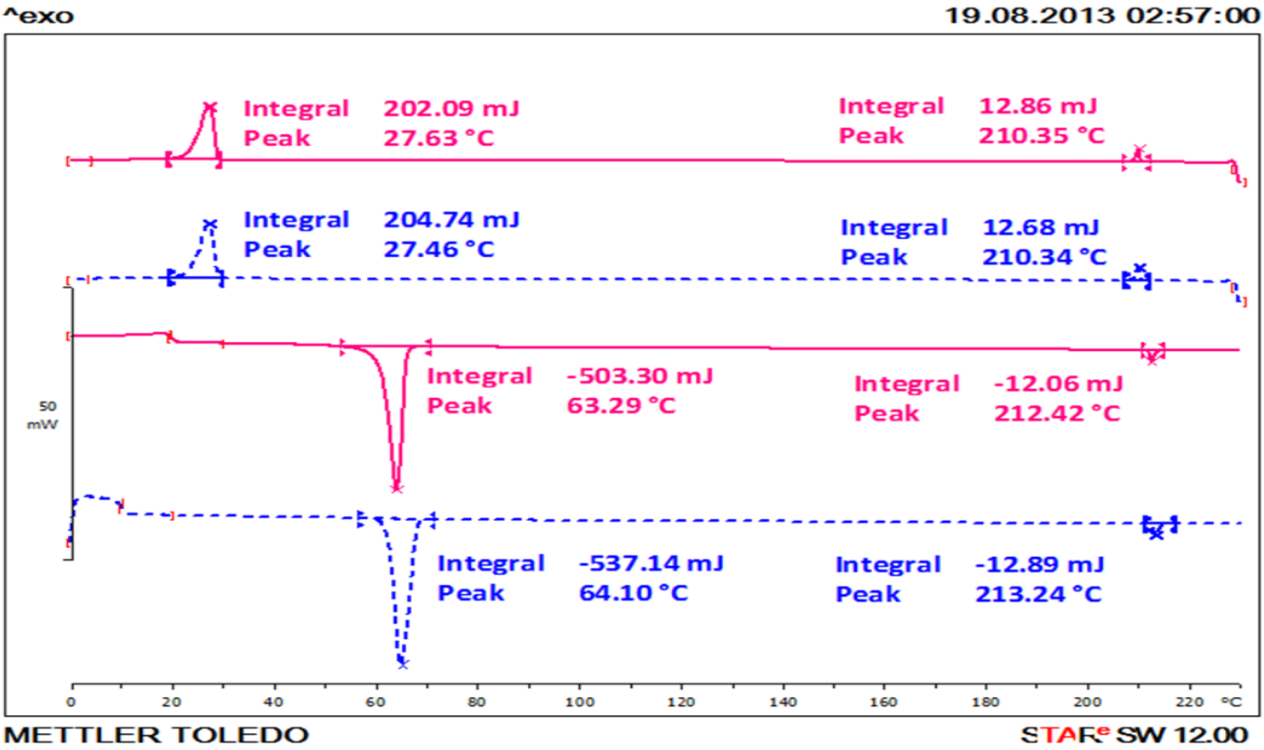

Figure S4 (b). DSC thermogram for $[C_1, EC_{14}\text{-Im}]BF_4$

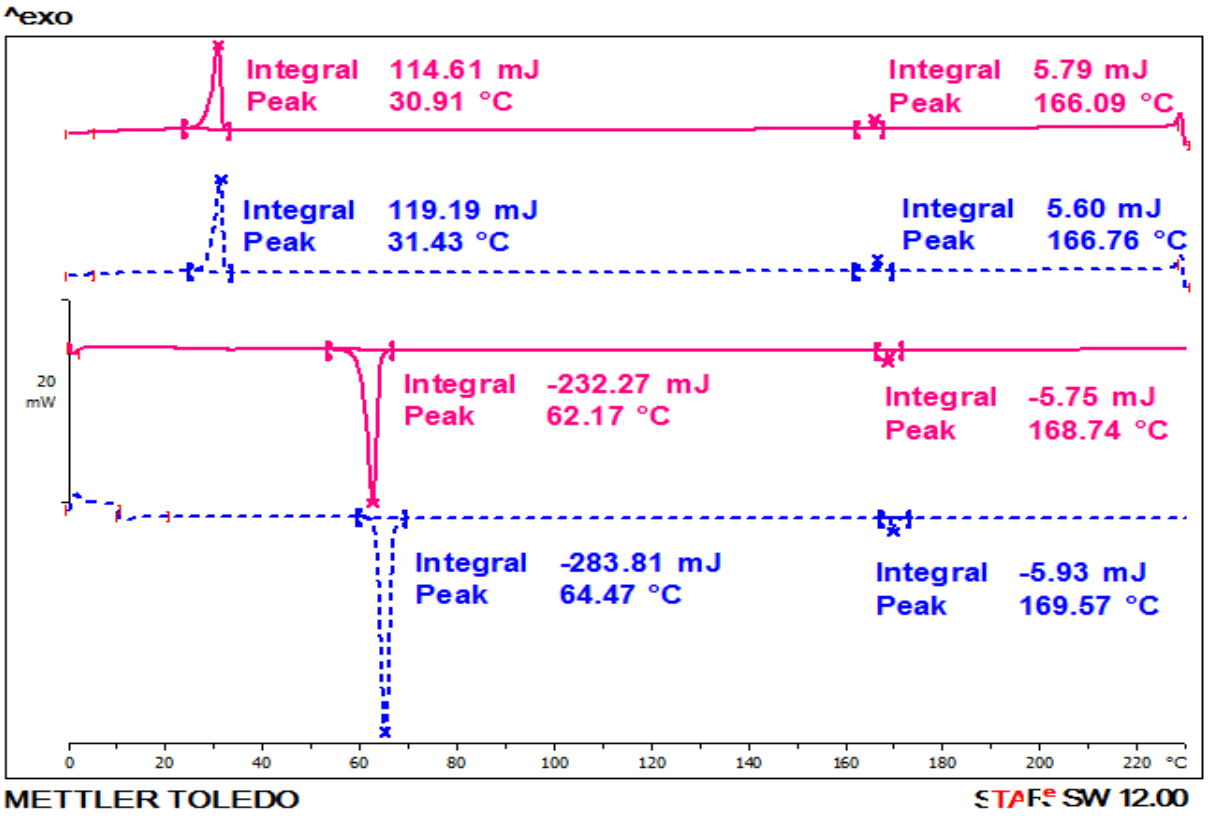

Figure S4 (c). DSC thermogram for $[C_1, EC_{14}\text{-Im}]PF_6$

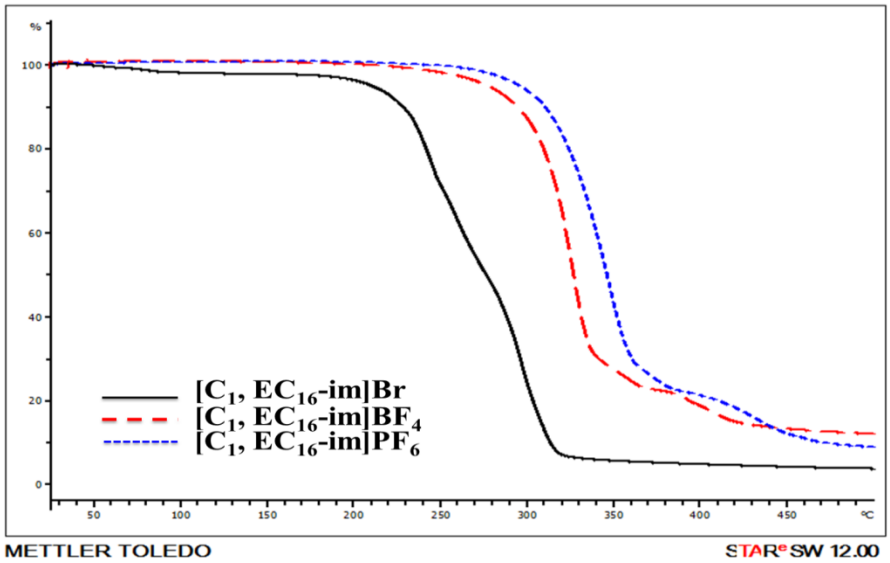

Figure S5. TGA curves for typical compound from each series

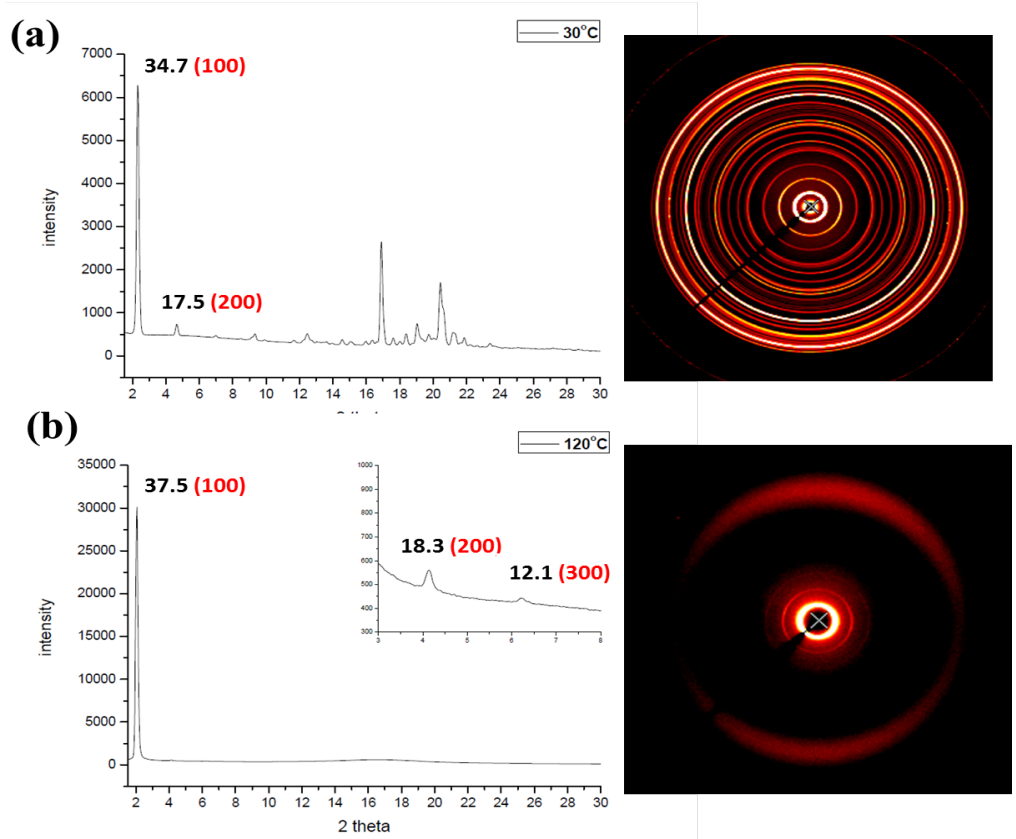

Figure S6. PXRD pattern of $[C_1, EC_{16}\text{-Im}]PF_6$ at crystal state (a), and at mesophase (b).

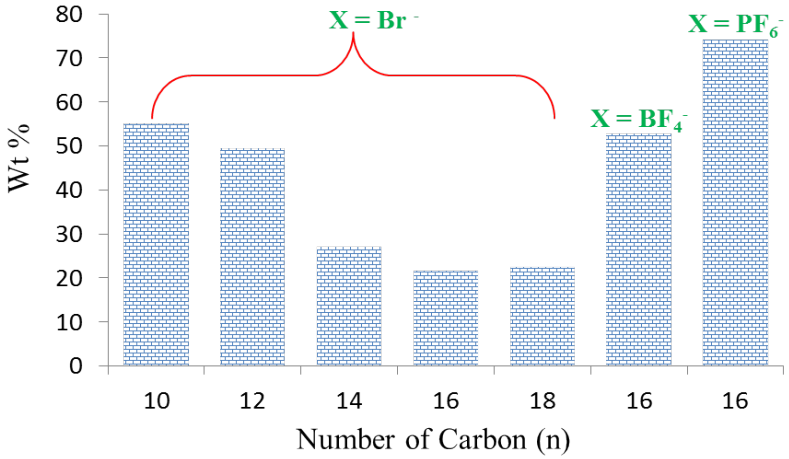

Figure S7. Schematic illustration of critical gel concentration of [C_1, EC_n-Im]Br (for n = 10, 12, 14, 16, and 18) and [C_1, EC_{16}-Im] X (for X = BF_4 and PF_6)

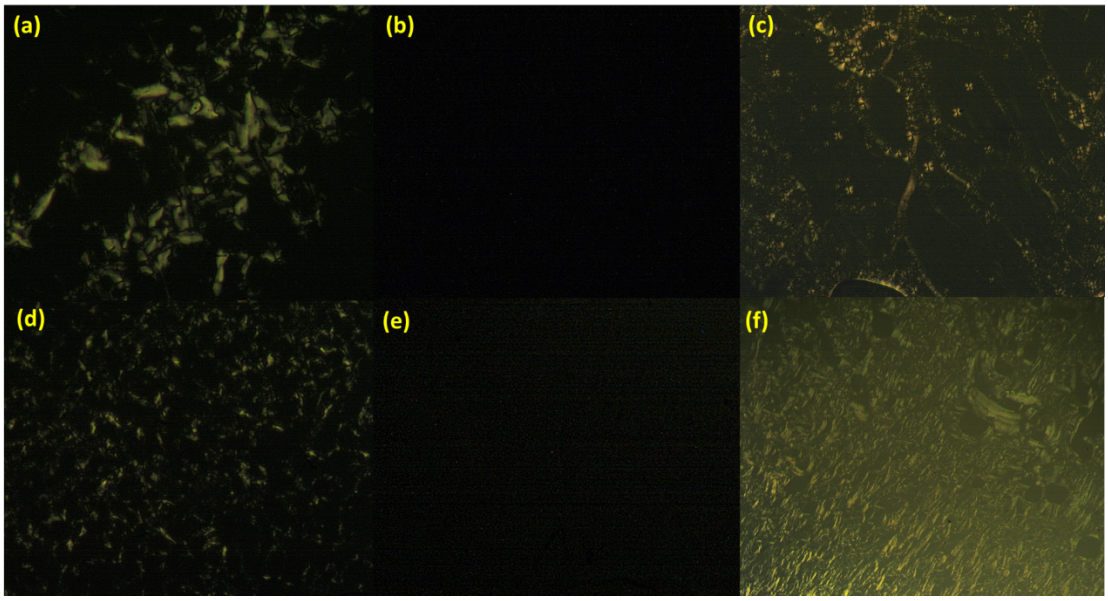

Figure S8. POM images of [C_1, EC_{14}-Im]BF_4 in $CHCl_3$ (a) at 45 wt% (lamellar SmA mesophase); (b) at 60 wt% (cubic phase); (c) at 60 wt% (lamellar LC gel); [C_1, EC_{14}-Im]PF_6 (d) at 60 wt % (lamellar SmA mesophase); (e) at 77 wt% (cubic phase); (f) at 77 wt% (lamellar LC gel).

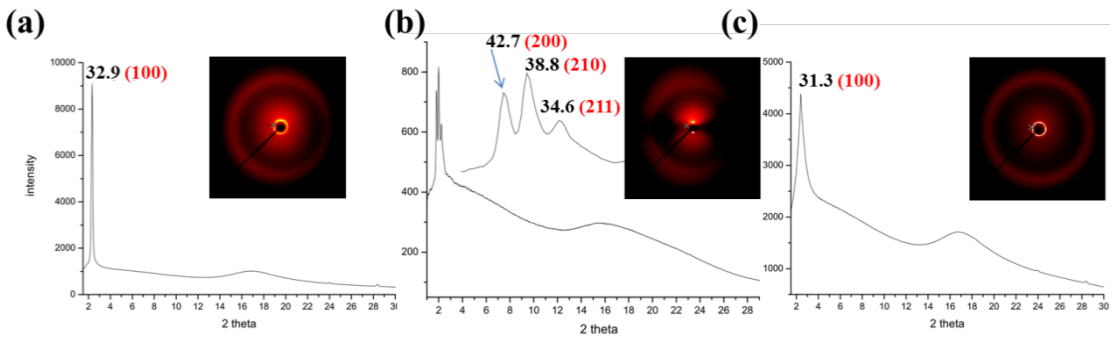

Figure S9. The PXRD data of [C_1, EC_{16}-Im]PF_6 salt in $CHCl_3$ solution at different salt gelling concentrations. (a) lamellar mesophase at 60 wt%; (b) cubic phase (Pm3n) before gelation at 77 wt%; (c) lamellar LC gel at 77 wt%.

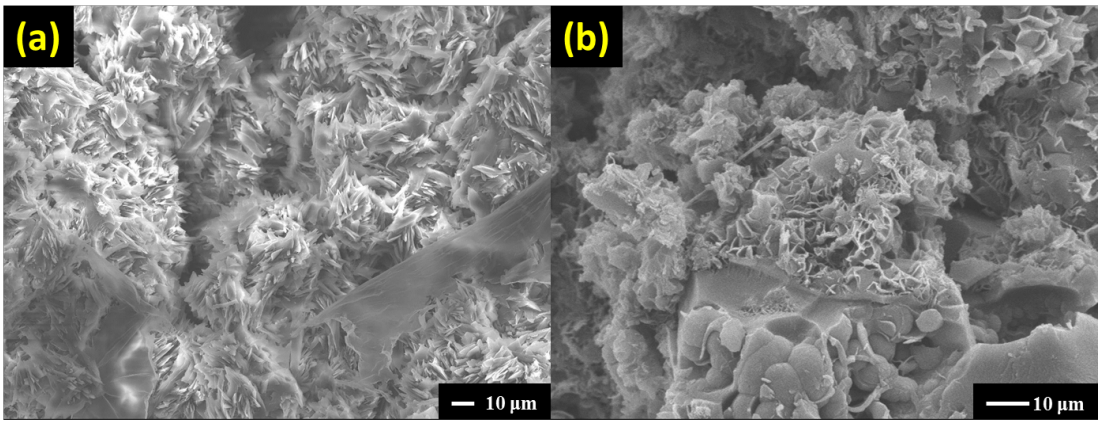

Figure S10. FE-SEM images of dried samples of (a): xerogel of [C_1, EC_{16}-Im]BF_4 and $CHCl_3$, (b):[C_1, EC_{16}-Im]PF_6 and $CHCl_3$.

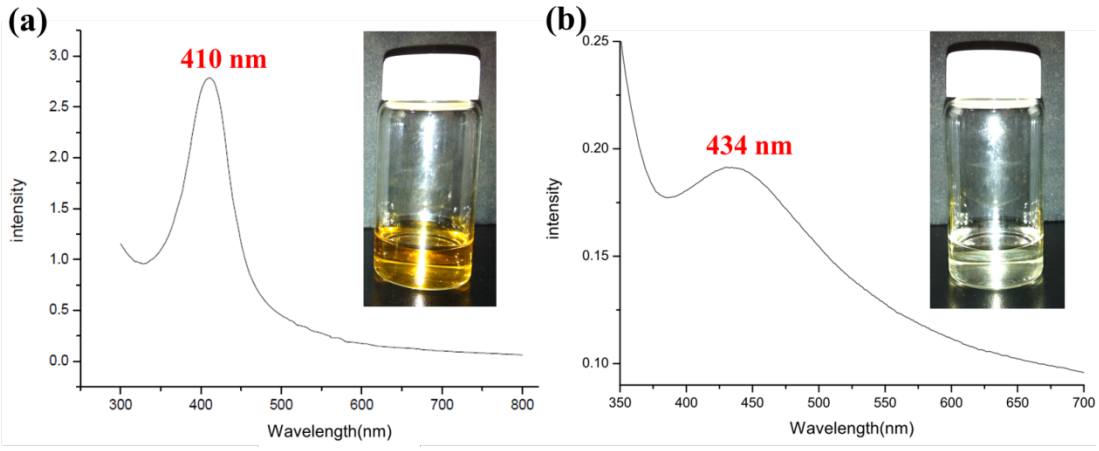

Figure S11. UV-Vis-images of Ag-NPs prepared in [C_1, EC_{16}-Im]Br LC gel; (a) taken after 2 days, and (b) taken after 5 days (sample images shows the disappearance of color after 3 days)

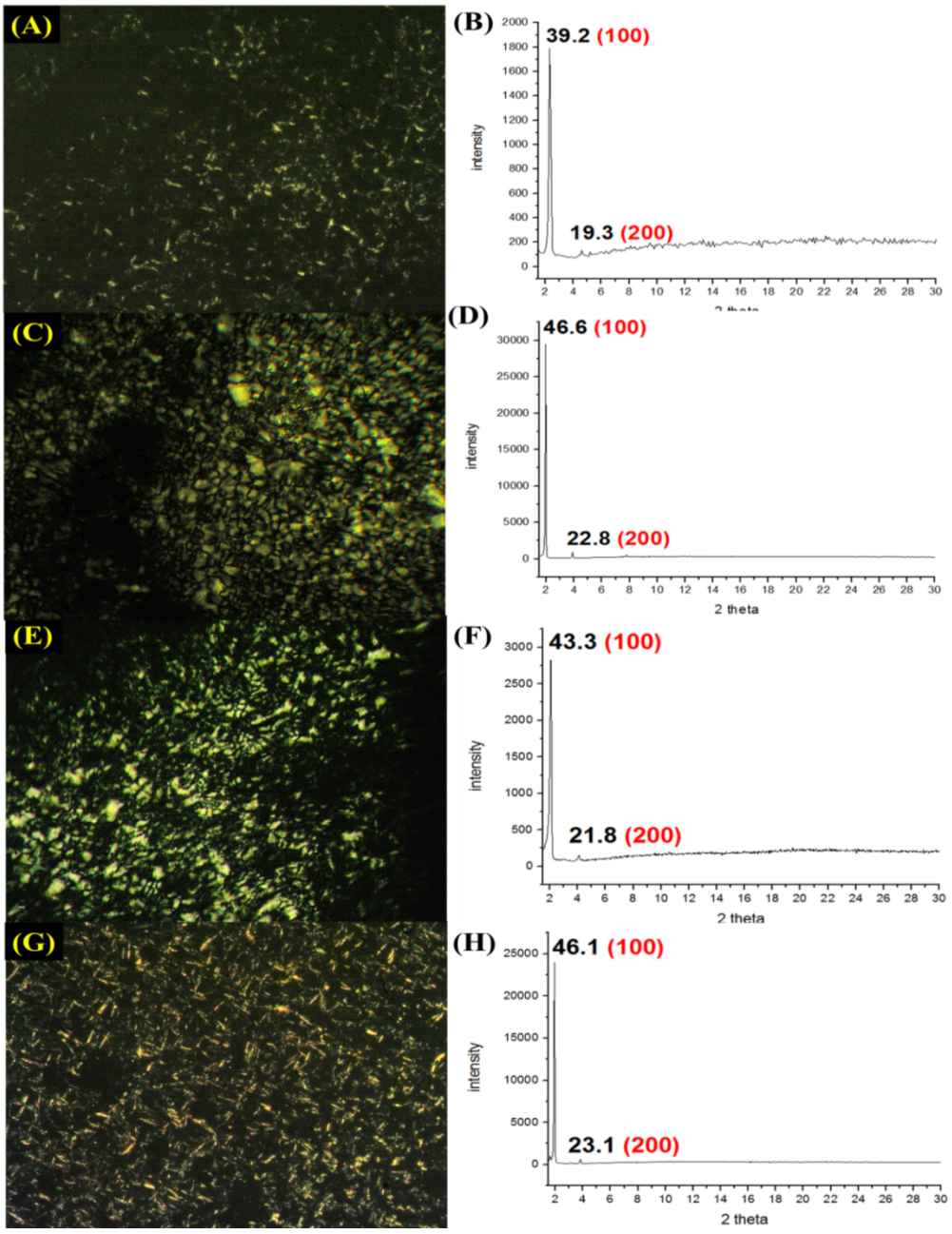

Figure S12. POM (broken fan like images), and PXRD (lamellar x-ray diffraction pattern) results: (A) and (B) are $AgNO_3$@[C_1, EC_{16}-Im]Br salt in $CHCl_3$ (20 wt%) at solution state; (C) and (D) are Ag-NPs@[C_1, EC_{16}-Im]Br salt in $CHCl_3$ (20 wt%) at solution state; (E) and (F) are $AgNO_3$@[C_1, EC_{16}-Im]Br salt in $CHCl_3$ (40 wt%) at gel state; (G) and (H) are AgNPs@[C_1, EC_{16}-Im]Br salt in $CHCl_3$ (40 wt%) at gel state.