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ELECTRONIC SUPPLEMENTARY MATERIAL

Towards room temperature ionic liquid crystals: linear versus tilted imidazolium phenylpyrimidines

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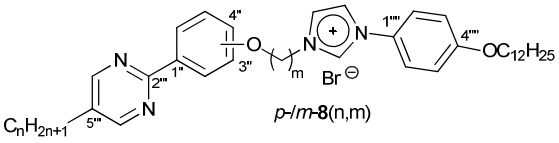
1. Experimental data

Compounds **10a-b**, **13a-c**, *p*-/*m*-**15**, *p*-**16**(8), *p*-**16**(10) and **18** are commercially available and compounds **9**,[1] **13d**,[2] **14**(8) and **14**(10),[3–5] *p*-/*m*-**19**,[6,7] *p*-**17**(8,4)[8] and *p*-**17**(10,8)[9] were prepared according to literature procedures. Atom numbering for the NMR assignment of compounds *p*-**8**(n,m) and *m*-**8**(n,m) is given in Scheme S1.



Scheme S1

For atom numbering of all other compounds see main paper.

1-{9-[3-(2-Dodecylpyrimidin-5-yl)phenoxy]nonyl}-3-methylimidazolium bromide (4). From **12** (0.13 mmol, 60.0 mg) and **13a** (1.50 mmol, 106 mg), 72 h; yield: 58.0 mg (72%). FT-IR (ATR): ν_{max}/cm^{-1} 3077 (m), 2923 (s), 2853 (m), 1602 (m), 1580 (m), 1543 (m), 1450 (s), 1372 (m), 1307 (m), 1210 (s), 1170 (m), 1048 (w), 1013 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, J = 6.9 Hz, CH_3), 1.20–1.51 (m, 28H, CH_2), 1.76–1.89 (m, 4H, CH_2), 1.90–1.97 (m, 2H, CH_2), 2.96–3.03 (m, 2H, 2'''-CH_2), 4.01 (t, 2H, J = 6.3 Hz, OCH_2), 4.13 (s 3H, NCH_3), 4.32 (t, 2H, J = 7.6 Hz, CH_2N), 6.94–6.99 (m, 1H, 6''-H), 7.05–7.08 (m, 1H, 2''-H), 7.10–7.14 (m, 1H, 4''-H), 7.20–7.26 (m, 2H, 4-H, 5-H), 7.40 (s, 1H, J = 8.2 Hz, 5''-H), 8.85 (s, 2H, 4'''-H, 6'''-H), 10.82 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 26.0, 26.3, 28.8, 28.9, 29.21, 29.27, 29.35, 29.46, 29.48, 29.55, 29.65, 29.67, 30.3, 31.9 (CH_2), 36.8 (NCH_3), 39.3 (2'''-CH_2), 50.3 (CH_2N), 68.1 (OCH_2), 113.3 (C-2''), 114.3 (C-6''), 119.1 (C-4''), 121.4, 122.9 (C-4, C-5), 130.4 (C-5''), 131.1 (C-5'''), 136.0 (C-3''), 138.6 (C-2), 155.1 (C-4''', C-6'''), 159.9 (C-1''), 170.5 (C-2''') ppm. MS (ESI): *m/z* = 547.4 $[M–Br]^+$, 465.4, 341.3, 207.2, 179.2, 165.1, 123.1, 83.1. HRMS (ESI): *m/z* calcd. for $C_{35}H_{55}N_4O^+$: 547.4370, found: 547.4365 $[M–Br]^+$. DSC: Cr 34 °C [16.6 kJ mol^{-1}] SmA 114 °C [0.3 kJ/mol] I (2nd heating), I 115 °C [-0.3 kJ/mol] SmA 17 °C [-20.4 kJ/mol] Cr (2nd cooling).

2-(3-Hydroxyphenyl)-3-(dimethylamino)acrolein (9). $POCl_3$ (102 mmol, 15.6 g) was added dropwise to dry DMF (165 mmol, 12.1 g) at 0 °C. After warming to room temperature (3-hydroxyphenyl)acetic acid (32.9 mmol, 5.00 g) was added portion-wise and the reaction mixture was stirred for 1 h at this temperature and for 5 h at 70 °C. After cooling to room temperature the reaction mixture was poured on crushed ice (150 mL) and sodium tetrafluoroborate (123 mmol, 13.5 g) was added immediately. After 1 h a yellow solid precipitated which was collected on a Buchner funnel and dried in vacuum to give 3-(3-

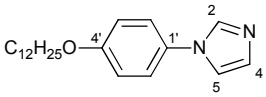
Hydroxyphenyl)-1,1,5,5-tetramethyl-1,5-diazapentadienium Tetrafluoroborate (7.98 g, 79%). δ_H (300 MHz, DMSO-d_6) 3.23 (s, 6H, 2 x CH_3), 3.23 (s, 6H, 2 x CH_3), 6.64–6.66 (m, 1H, 2'-H), 6.68–6.74 (m, 1H, 6'-H), 6.80–6.86 (m, 1H, 4'-H), 7.23 (t, 1H, *J* = 7.9 Hz, 5'-H), 7.64 (s, 1H, 3-H), 9.74 (s, 1H, 1-H) ppm. δ_C (75 MHz, DMSO-d_6) 48.4 (CH_3), 105.0 (C-2), 115.7 (C-2'), 118.7 (C-4'), 122.8 (C-6'), 129.5 (C-5'), 133.6 (C-1'), 157.1 (C-1, C-3'), 162.7 (C-3) ppm. The NMR data are in good agreement with those of the corresponding perchlorate.[1]

A suspension of the tetrafluoroborate (6.50 mmol, 2.00 g) and Na_2CO_3 (12.3 mmol, 1.3 g) in ethanol/water (1:1, 10 mL) was heated to reflux for 2 h. After cooling to room temperature the mixture was diluted with water (10 mL) and glacial acetic acid was added until the solution turned yellow. The solution was concentrated in vacuum to one quarter of the original volume and stored in the refrigerator for 12 h. The precipitate was collected on a funnel and dried in vacuum to give **9** (546 mg, 44%). δ_H (300 MHz, DMSO-d_6) 3.37 (s, 6H, 2 x CH_3), 6.46–6.48 (m, 1H, 2'-H), 6.48–6.51 (m, 1H, 6'-H), 6.60–6.64 (m, 1H, 4'-H), 7.08 (t, 1H, *J* = 7.9 Hz, 5'-H), 7.09 (s, 1H, 3-H), 8.91 (s, 1H, 1-H) ppm. δ_C (75 MHz, DMSO-d_6) 113.2 (C-2'), 114.2 (C-2), 118.0 (C-4'), 121.8 (C-6'), 128.1 (C-5'), 156.4 (C-3, C-3'), 159.1 (C-1'), 187.8 (C-1) ppm. The spectroscopic data are in good agreement with those reported in literature.[1]

3-(2-Dodecyl pyrimidin-5-yl)phenol (11). A solution of 2-(3-hydroxyphenyl)-3-(dimethylamino)acrolein **9** (1.42 mmol, 272 mg) and dodecanamidine **10a** (2.10 mmol, 271 mg) in pyridine (3.00 mL) was stirred at 70 °C for 6 h. After cooling to room temperature and dilution with H_2O (4 mL), the reaction mixture was adjusted to pH = 1 with half conc. HCl and extracted with Et_2O (3 × 6 mL). The combined organic layers were washed with H_2O (4 mL), dried (Na_2SO_4) and the solvent was removed under reduced pressure. The residue was purified by chromatography on SiO_2 (hexanes/EtOAc 1 : 1) to give **11** (92.0 mg, 19%) as a colourless solid. R_f = 0.47 (hexanes/EtOAc 1 : 1). IR (ATR): ν_{max}/cm^{-1} 2953 (m), 2919 (s), 2851 (s), 1600 (s), 1543 (w), 1453 (s), 1401 (m), 1240 (m), 1222 (s), 1032 (w), 837 (m), 722 (s), 657 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 1.17–1.46 (m, 18H, CH_2), 1.80–1.92 (m, 2H, CH_2), 2.97–3.05 (m, 2H, 2-CH_2), 6.95–7.00 (m, 1H, 6'-H), 7.06–7.09 (m, 1H, 2'-H), 7.11–7.15 (m, 1H, 4'-H), 7.39 (t, 1H, *J* = 8.2 Hz, 5'-H), 7.53 (s, br, 1H, OH), 8.94 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 28.9, 29.36, 29.39, 29.43, 29.52, 29.64, 29.66, 31.9 (CH_2), 39.0 (2-CH_2), 113.8 (C-6'), 116.2 (C-2'), 118.7 (C-4'), 130.9 (C-5'), 131.4 (C-5), 135.7 (C-3'), 154.9 (C-4, C-6), 157.2 (C-1'), 170.4 (C-2) ppm. MS (EI): *m/z* (%) = 340.2 (60) $[M]^+$, 297.2 (3), 269.1 (2), 241.1 (6), 213.1 (6), 199.1 (28), 186.0 (100),

157.1 (2), 118.0 (3), 89.0 (1), 69.1 (1), 43.0 (2). HRMS (ESI): *m/z* calcd. for $C_{22}H_{33}N_2O^+$: 341.2587, found: 341.2581 $[MH]^+$.

1-[4'-(Dodecyloxy)phenyl]-imidazole (13d). In a modification of literature procedures[2] 1-bromo-4-dodecyloxybenzene (10.3 mmol, 3.50 g) was dissolved in dry, degassed DMF (35 ml) under a nitrogen atmosphere and cesium carbonate (10.3 mmol, 3.36 g), imidazole (14.4 mmol, 980 mg) and copper(I)iodide (2.06 mmol, 392 mg) were added subsequently. The reaction mixture was heated to 60 °C for 5 d and after cooling to room temperature quenched with water (20 ml). The precipitates were separated by filtration and the filtrate was extraxcted with CH_2Cl_2 (3 x 25 mL). The combined organic extracts were dried over Na_2SO_4 and the solvent was removed under reduced pressure. The residue was purified by chromatography on SiO_2 (hexanes/EtOAc 5 : 1) to give **13d** (2.37 g, 70%) as a colourless solid.



Found: C, 76.65; H, 9.72; N, 8.46%. $C_{21}H_{32}N_2O$ requires C, 76.78; H, 8.82; N, 8.53%; M, 328.49. FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2853 (m), 1612 (m), 1519 (s), 1468 (m), 1299 (m), 1245 (s), 1176 (w), 1057 (s), 904 (s), 830 (m), 727 (s), 652 (m). δ_H (250 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.19–1.40 (m, 16H, CH_2), 1.42–1.50 (m, 2H, CH_2), 1.75–1.86 (m, 2H, CH_2), 3.98 (t, 2H, *J* = 6.6 Hz, OCH_2), 6.93–7.02 (m, 2H, 3'-H, 5'-H), 7.16–7.26 (s, br, 2H, 2-H, 4-H), 7.26–7.31 (m, 2H, 2'-H, 6'-H), 7.78 (s, 1H, 5-H) ppm. δ_C (63 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 26.0, 29.2, 29.35, 29.38, 29.57, 29.60, 29.64, 29.7, 31.9 (CH_2), 68.5 (OCH_2), 115.5 (C-3', C-5'), 118.2 (C-5), 123.2 (C-2', C-6'), 128.5 (C-1'), 130.6 (C-4), 135.5 (C-2), 158.5 (C-4') ppm. MS (EI): *m/z* (%) = 328.2 (64) $[M]^+$, 173.0 (3), 160.0 (100), 133.0 (12), 106.0 (6), 85.1 (1), 71.1 (3), 57.0 (5), 43.0 (6). HRMS (ESI): *m/z* calcd. for $C_{21}H_{23}N_2O^+$: 329.2587, found: 329.2592 $[MH]^+$. Analytic data are in good agreement with those reported in literature.[2]

General procedure for the preparation of 2-(methoxymethylene) alkanals 14(n).[3] Dry DMF (168 mmol) was added dropwise under vigorous stirring and cooling in an ice-bath to $POCl_3$ (154 mmol). After complete addition the reaction mixture was warmed to 40 °C and the corresponding 1,1-dimethoxyalkane (70 mmol) was added dropwise using a dropping funnel. Afterwards the reaction mixture was heated to 60 °C for 2 h, cooled to room temperature and crushed ice (40 g) was added. Finally the mixture was stored in the refrigerator for 12 h and the pH of the solution was adjusted to 8 by the addition of potassium carbonate. The precipitate was separated by filtration and the filtrate was extracted with CH_2Cl_2 (3 x 100 mL). The combined organic extracts were dried over Na_2SO_4 and the solvent

was removed under reduced pressure. The residue was purified by chromatography on SiO_2 using mixtures of hexanes and EtOAc to give **14**(n) as yellow oils.

2-(Methoxymethylene)decanal (14(8)).[4] From 1,1-dimethoxydecane (45.7 mmol, 9.19 g); yield: 4.13 g (46%), yellow oil. R_f = 0.34 (hexanes/ EtOAc 10 : 1). FT-IR (ATR): ν_{max}/cm^{-1} 2924 (m), 2854 (m), 2728 (w), 1670 (m), 1637 (s), 1458 (w), 1385 (w), 1304 (w), 1249 (m), 1213 (m), 1145 (m), 1110 (w), 1019 (w), 981 (w), 889 (w). δ_H (250 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.6 Hz, CH_3), 1.18–1.35 (m, 12H, CH_2), 2.13–2.26 (m, 2H, CCH_2), 3.94 (s, 3H, OCH_3), 6.86 [s, 0.6H, (*E*)-*H*C(OCH_3)], 6.93 [s, 0.4H, (*Z*)-*H*C(OCH_3)], 9.19 (s, 1H, CHO) ppm. δ_C (63 MHz, $CDCl_3$) 14.1 (CH_3), 15.3, 21.5, 22.7, 28.0, 31.9 (CH_2), 62.0 (OCH_3), 124.9 (C-2), 168.1, 169.2 [(*E*)-, (*Z*)-*C*(OCH_3)], 191.7 (CHO) ppm. MS (ESI): *m/z* (%) = 199.17 $[MH]^+$, 185.2, 123.1, 107.1. HRMS (ESI): *m/z* calcd. for $C_{12}H_{22}O_2^+$: 198.1620, found: 198.1614 $[M]^+$.

2-(Methoxymethylene)dodecanal (14(10)).[5] From 1,1-dimethoxydodecane (67.0 mmol, 15.3 g); yield: 0.39 g (3%), yellow oil. R_f = 0.42 (hexanes/ EtOAc 5 : 1). FT-IR (ATR): ν_{max}/cm^{-1} 2922 (s), 2853 (m), 2729 (w), 1670 (m), 1638 (s), 1465 (w), 1386 (w), 1304 (w), 1251 (m), 1215 (m), 1146 (m), 1110 (w), 1019 (w), 983 (w). δ_H (250 MHz, $CDCl_3$) 0.89 (t, 3H, *J* = 6.6 Hz, CH_3), 1.18–1.36 (m, 16H, CH_2), 2.15–2.25 (m, 2H, CCH_2), 3.95 (s, 3H, OCH_3), 6.86 [s, 0.33H, (*E*)-*H*C(OCH_3)], 6.93 [s, 0.67H, (*Z*)-*H*C(OCH_3)], 9.20 (s, 1H, CHO) ppm. δ_C (63 MHz, $CDCl_3$) 14.1 (CH_3), 15.3, 21.5, 22.7, 28.0, 31.9 (CH_2), 62.0 (OCH_3), 124.8 (C-2), 168.1, 169.2 [(*E*),(*Z*)-*C*(OCH_3)], 191.7 (CHO) ppm. MS (ESI): *m/z* (%) = 227.2 $[MH]^+$, 213.2. HRMS (ESI): *m/z* calcd. for $C_{14}H_{27}O_2^+$: 227.2006, found: 227.2003 $[MH]^+$.

2-(Methoxymethylene)tetraecanal (14(12)). From 1,1-dimethoxytetradecane (70.0 mmol, 18.1 g); yield: 3.99 g (22%), yellow oil. R_f = 0.25 (hexanes/ EtOAc 5 : 1). FT-IR (ATR): ν_{max}/cm^{-1} 2922 (s), 2853 (m), 1730 (w), 1672 (w), 1638 (m), 1465 (w), 1379 (w), 1303 (w), 1215 (m), 1145 (w), 1110 (w), 1019 (w). δ_H (250 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.5 Hz, CH_3), 1.19–1.35 (m, 20H, CH_2), 2.14–2.25 (m, 2H, CCH_2), 3.94 (s, 3H, OCH_3), 6.85 [s, 0.47H, (*E*)-*H*C(OCH_3)], 6.93 [s, 0.53H, (*Z*)-*H*C(OCH_3)], 9.19 (s, 1H, CHO) ppm. δ_C (63 MHz, $CDCl_3$) 14.1 (CH_3), 15.3, 21.5, 22.7, 28.0, 31.9 (CH_2), 62.1 (OCH_3), 124.8 (C-2), 168.3, 169.4 [(*E*),(*Z*)-*C*(OCH_3)], 191.8 (CHO) ppm. MS (ESI): *m/z* (%) = 255.2 $[MH]^+$, 241.2, 212.2. HRMS (ESI): *m/z* calcd. for $C_{16}H_{31}O_2^+$: 255.2319, found: 255.2315 $[M]^+$.

General procedure for the preparation of *p*- and *m*-hydroxybenzamidine hydrochlorides *p*-/*m*-15. In a modification of literature procedures[6,7] dry hydrogen chloride was passed through a solution of 10 g (83.9 mmol) of *p*- or *m*-hydroxybenzonitrile in dry methanol (100 mL) for 2 h. In a strongly exothermic reaction a yellow solid precipitated. After cooling to

room temperature the solvent was evaporated and the residue was suspended in dry ethanol (100 mL). Dry ammonia was passed through this suspension for 2 h which therefore changed her color from yellow to pink. Finally the reaction mixture was heated to 60 °C for another 2 h. The solvent was removed in vacuo and the brown residue was dissolved in methanol (200 mL). Afterwards dry diethyl ether was added to force the precipitation of ammonium chloride, which was removed by filtration. The filtrate was evaporated to dryness and the pale brown residue used without further purification.

***p*-Hydroxybenzamidine hydrochloride (*p*-15).**[6] From *p*-hydroxybenzonitrile (83.9 mmol, 10.0 g); yield: 10.1 g (69%). FT-IR (ATR): ν_{max}/cm^{-1} 3144 (br), 2364 (w), 1668 (s), 1609 (s), 1487 (s), 1439 (m), 1287 (m), 1235 (m), 1183 (m), 1125 (w), 970 (w), 845 (m), 746 (w), 669 (w), 631 (w), 593 (w), 545 (w), 538 (w), 530 (w). δ_H (250 MHz, $CDCl_3$) 6.90–7.01 (m, 2H, 3-H, 5-H), 7.63–7.78 (m, 2H, 2-H, 6-H) ppm. δ_C (63 MHz, $CDCl_3$) 117.2, 119.4, 131.2, 164.7, 168.4 ppm. The spectroscopic data are in good agreement with those reported in literature.[6]

***m*-Hydroxybenzamidine hydrochloride (*m*-15).**[7] From *m*-hydroxybenzonitrile (83.9 mmol, 10.0 g); yield: 11.8 g (81%). FT-IR (ATR): ν_{max}/cm^{-1} 3126 (br), 2359 (w), 1677 (s), 1589 (m), 1533 (w), 1479 (m), 1445 (m), 1342 (w), 1273 (m), 1235 (m), 1095 (w), 999 (w), 929 (w), 874 (w), 796 (w), 716 (w), 678 (w). δ_H (250 MHz, $CDCl_3$) 7.08–7.30 (m, 3H, 2-H, 4-H, 6-H), 7.42 (t, 1H, J = 7.9 Hz, 5-H) ppm. δ_C (63 MHz, $CDCl_3$) 115.6, 119.6, 122.1, 130.9, 131.7, 159.6, 168.7 ppm. MS (ESI): *m/z* = 137.1 $[M\text{-}Cl]^+$, 120.0. HRMS (ESI): *m/z* calcd. for $C_7H_9N_2O^+$: 137.0709, found: 137.0699 $[M\text{-}Cl]^+$.

General procedure for the preparation of 4- and 3-(5-alkylpyrimidin-2-yl)phenols *p*-/*m*-16(n). To a solution of ***p*-15** or ***m*-15** (1.50 mmol) in abs. methanol (3 mL) under inert gas was added the respective 2-(methoxymethylidene)alkanal **14**(n) (1.00 mmol) and a 2 M NaOMe solution (6.8 mL, 13.6 mmol). A light orange colour was observed and NaCl precipitated. After heating to reflux for 2 h, the reaction mixture was stirred for further 12 h at room temperature. The precipitated salt was filtered off and the filtrate was concentrated under reduced pressure. The residue was purified by chromatography on SiO_2 (hexanes/EtOAc 1 : 1) to give the products as colourless solid.

4-(5-Octylpyrimidin-2-yl)phenol (*p*-16(8)).[10] From ***p*-15** (6.81 mmol, 1.18 g), **18**(8) (4.54 mmol, 900 mg) and NaOMe (2 M, 7.00 mL, 14.0 mmol) in MeOH (14.0 mL); yield: 572 mg (44%). R_f = 0.50 (hexanes/EtOAc 5 : 1). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, J = 6.6 Hz, CH_3), 1.20–1.41 (m, 12H, CH_2), 1.60–1.68 (m, 2H, CH_2), 2.57–2.63 (m, 2H, 5-CH_2), 6.89–6.95 (m, 2H, 2'-H, 6'-H), 8.28–8.34 (m, 2H, 3'-H, 5'-H), 8.58 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.6, 29.2, 29.29, 29.32, 29.56, 30.1, 30.8, 31.9 (CH_2), 115.5

(C-2', C-6'), 129.7 (C-4'), 130.6 (C-3', C-5'), 132.3 (C-5), 157.0 (C-4, C-6), 158.7 (C-1'), 162.5 (C-2) ppm. Spectroscopic data are in good agreement with those reported in literature.[10]

4-(5-Decylpyrimidin-2-yl)phenol (*p*-16(10)).[5a] From *p*-**15** (7.29 mmol, 1.26 g), **18**(10) (4.86 mmol, 1.10 g) and NaOMe (2 M, 7.50 mL, 15.0 mmol) in MeOH (15.0 mL); yield: 744 mg (49%). R_f = 0.52 (hexanes/EtOAc 5 : 1). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 1.19–1.41 (m, 14H, CH_2), 1.57–1.68 (m, 2H, CH_2), 2.56–2.64 (m, 2H, 5-CH_2), 6.81–6.89 (m, 2H, 2'-H, 6'-H), 8.02 (s, br, 1H, OH), 8.16–8.25 (m, 2H, 3'-H, 5'-H), 8.60 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.7, 29.1, 29.29, 29.32, 29.51, 29.56, 30.1, 30.8, 31.9 (CH_2), 115.7 (C-2', C-6'), 129.4 (C-4'), 129.8 (C-3', C-5'), 132.4 (C-5), 157.0 (C-4, C-6), 158.8 (C-1'), 162.5 (C-2) ppm.

4-(5-Dodecylpyrimidin-2-yl)phenol (*p*-16(12)). From *p*-**15** (8.87 mmol, 1.53 g), **18**(12) (5.90 mmol, 1.50 g) and NaOMe (2 M, 13.5 mL, 27.0 mmol) in MeOH (20 mL); yield: 1.08 g (54%). R_f = 0.54 (hexanes/ EtOAc 5 : 1). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 1.18–1.39 (m, 18H, CH_2), 1.58–1.69 (m, 2H, CH_2), 2.56–2.64 (m, 2H, 5-CH_2), 6.82–6.89 (m, 2H, 2'-H, 6'-H), 8.17–8.24 (m, 2H, 3'-H, 5'-H), 8.49 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 28.7, 29.0, 29.27, 29.34, 29.47, 29.61, 29.64, 30.1, 30.8, 31.9 (CH_2), 115.7 (C-2', C-6'), 129.3 (C-4'), 129.8 (C-3', C-5'), 132.4 (C-5), 157.0 (C-4, C-6), 158.8 (C-1'), 162.5 (C-2) ppm.

3-(5-Dodecylpyrimidin-2-yl)phenol (*m*-16(12)). From *m*-**15** (5.31 mmol, 917 mg), **18**(12) (3.54 mmol, 900 mg) and NaOMe (2 M, 8.00 mL, 16.0 mmol) in MeOH (11.0 mL); yield: 759 mg (63%). R_f = 0.52 (hexanes/EtOAc 5 : 1). Found: C, 76.44; H, 9.35; N, 7.19%. $C_{22}H_{32}N_2O$ requires C, 77.60; H, 9.47; N, 8.23%; M, 340.50. FT-IR (ATR): ν_{max}/cm^{-1} 3152 (br), 2921 (s), 2849 (m), 1723 (w), 1701 (w), 1597 (m), 1547 (m), 1451 (m), 1432 (s), 1402 (w), 1325 (m), 1242 (m), 1082 (w), 998 (w), 936 (w), 898 (w), 844 (w), 776 (w), 725 (w), 684 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.21–1.39 (m, 18H, CH_2), 1.59–1.69 (m, 2H, CH_2), 2.57–2.69 (m, 2H, 5-CH_2), 6.93–6.98 (m, 1H, 6'-H), 7.33 (t, 1H *J* = 7.9 Hz, 5'-H), 7.89–7.96 (m, 2H, 2'-H, 4'-H), 8.62 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.6, 29.1, 29.2, 29.27, 29.29, 29.34, 29.47, 29.61, 29.64, 30.2, 30.8 , 31.9 (CH_2), 114.8 (C-2'), 117.9 (C-6'), 120.3 (C-4'), 130.0 (C-5'), 133.4 (C-3'), 138.9 (C-5), 156.5 (C-1'), 157.1 (C-4, C-6), 162.2 (C-2) ppm. MS (EI): *m/z* = 340.3 (100) $[M]^+$, 352.2 (10), 311.2 (20), 283.2 (17), 255.2 (14), 227.1 (21), 213.1 (13), 199.1 (74), 186.1 (62), 152.0 (60), 121.0 (96), 93.0 (24), 81.1 (4), 65.0 (10), 43.1 (15). HRMS (ESI): *m/z* calcd. for $C_{22}H_{32}N_2O^+$: 341.2587, found: 341.2494 $[M]^+$.

General procedure for the preparation of [(bromoalkyl)oxy]alkylpyrimidines 12 and *p*-/*m*-17(n,m). To a solution of the respective alcohol **11**, *p*-**16**(n) or *m*-**16**(n) (1 mmol) in DMSO (5 mL) was added finely powdered potassium hydroxide (3.00 mmol) and the reaction mixture stirred at room temperature for 10 min. After addition of the respective α,ω-dibromoalkane (1.50 mmol for **11** and 2.00 mmol for *p*-**16**(n) and *m*-**16**(n)), the reaction mixture was stirred at room temperature for 12 h followed by addition of H_2O (10 mL) and CH_2Cl_2 (10 mL). The layers were separated, and the aqueous layer was extracted with CH_2Cl_2 (2 × 20 mL). The combined organic layers were dried (Na_2SO_4) and the solvent was removed under reduced pressure. The residue was purified by chromatography on SiO_2 (hexanes/ EtOAc 10 : 1).

5-{3-[(9-Bromononyl)oxy]phenyl}-2-dodecylpyrimidine (12). From **11** (0.21 mmol, 70.0 mg) and $Br(CH_2)_9Br$ (0.32 mmol, 91.0 mg); yield: 65.0 mg (66%), colourless solid. R_f = 0.45 (hexanes/ EtOAc 5 : 1); mp 75 °C. FT-IR (ATR): ν_{max}/cm^{-1} 2924 (s), 2853 (s), 1582 (m), 1542 (m), 1449 (s), 1371 (m), 1209 (s), 1047 (w), 1014 (w), 779 (m). MS (EI): *m*/*z* (%) = 546.3 $[M+H]^+$ (100), 501.2 (2), 447.2 (3), 392.1 (20), 353.2 (3), 311.2 (18), 269.1 (2), 186.0 (9), 157.0 (3), 118.0 (1), 83.1 (1), 55.0 (6). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.19–1.53 (m, 28H, CH_2), 1.75–1.91 (m, 6H, CH_2), 2.95–3.07 (m, 2H, 2-CH_2), 3.41 (t, 2H, *J* = 6.9 Hz, CH_2Br), 4.01 (t, 2H, *J* = 6.3 Hz, OCH_2), 6.93–6.98 (m, 1H, 6'-H), 7.06–7.08 (m, 1H, 2'-H), 7.10–7.14 (m, 1H, 4'-H), 7.40 (t, 1H, *J* = 7.9 Hz, 5'-H), 8.85 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 26.0, 28.10, 28.7, 28.8, 29.2, 29.26, 29.35, 29.45, 29.48, 29.55, 29.65, 29.67, 31.9, 32.8, 34.0 (CH_2), 39.3 (2-CH_2), 68.1 (OCH_2), 113.3 (C-2'), 114.3 (C-6'), 119.0 (C-4'), 130.4 (C-5'), 131.1 (C-5), 136.0 (C-3'), 155.0 (C-4, C-6), 159.8 (C-1'), 170.5 (C-2) ppm. HRMS (ESI): *m*/*z* calcd. for $C_{31}H_{50}{}^{79}BrN_2O^+$: 545.3101; found: 545.3092 $[M+H]^+$; calcd. for $C_{31}H_{50}{}^{81}BrN_2O^+$: 547.3086; found: 547.3096 $[M+H]^+$.

2-[4-(4-Bromobutoxy)phenyl]-5-octylpyrimidine (*p*-17(8,4)).[8] From *p*-**16**(8) (0.88 mmol, 251 mg) and $Br(CH_2)_4Br$ (1.77 mmol, 381 mg); yield: 208 mg (56%), colourless solid. R_f = 0.35 (hexanes/ethyl acetate 20 : 1). FT-IR (ATR): ν_{max}/cm^{-1} 2926 (w), 2855 (w), 1608 (w), 1585 (w), 1542 (w), 1432 (w), 1252 (w), 1169 (w), 903 (s), 846 (w), 798 (w), 723 (s), 650 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.20–1.40 (m, 10H, CH_2), 1.60–1.68 (m, 2H, CH_2), 1.94–2.02 (m, 2H, CH_2), 2.05–2.13 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5-CH_2), 3.50 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.06 (t, 2H, *J* = 6.0 Hz, OCH_2), 6.94–7.00 (m, 2H, 2'-H, 6'-H), 8.32–8.38 (m, 2H, 3'-H, 5'-H), 8.57 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.6, 27.9, 29.0, 29.2, 29.3, 29.5, 30.2, 30.8, 31.8, 33.4 (CH_2), 66.9 (OCH_2), 114.4 (C-2', C-6'), 129.4 (C-3', C-5'), 130.4 (C-4'), 132.2 (C-5), 157.0 (C-4, C-6), 160.9 (C-1'), 162.4

(C-2) ppm. DSC: Cr 41 °C [25.2 kJ/mol] SmA 52 °C [2.7 kJ/mol] I (2nd heating); I 59 °C [-0.5 kJ/mol] N 56 °C [-0.5 kJ/mol] SmA 3 °C [-13.0 kJ/mol] Cr (2nd cooling).

2-[4-(4-Bromobutoxy)phenyl]-5-decylpyrimidine (*p*-17(10,4)).

From *p*-**16**(10) (3.78 mmol, 1.18 g) and $Br(CH_2)_4Br$ (5.66 mmol, 1.22 g); yield: 1.47 g (87%), colourless solid. R_f = 0.36. Found: C, 64.57; H, 7.88; N, 6.05%. $C_{24}H_{35}BrN_2O$ requires C, 64.42; H, 7.88; N, 6.26%; M, 447.45. FT-IR (ATR): ν_{max}/cm^{-1} 2925 (w), 2853 (w), 2016 (w), 1608 (w), 1585 (w), 1542 (w), 1514 (w), 1467 (w), 1431 (s), 1328 (w), 1253 (m), 1169 (w), 1107 (w), 1043 (w), 909 (w), 846 (w), 798 (m), 733 (w), 654 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.21–1.40 (m, 14H, CH_2), 1.59–1.68 (m, 2H, CH_2), 1.93–2.02 (m, 2H, CH_2), 2.05–2.13 (m, 2H, CH_2), 2.55–2.63 (m, 2H, 5-CH_2), 3.50 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.07 (t, 2H, *J* = 6.0 Hz, OCH_2), 6.93–7.02 (m, 2H, 2'-H, 6'-H), 8.31–8.39 (m, 2H, 3'-H, 5'-H), 8.60 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 27.9, 29.0, 29.30, 29.35, 29.45, 29.53, 29.57, 30.2, 30.8, 31.9, 33.4 (CH_2), 66.9 (OCH_2), 114.4 (C-2', C-6'), 129.4 (C-3', C-5'), 130.4 (C-4'), 132.2 (C-5), 157.0 (C-4, C-6), 160.9 (C-1'), 162.4 (C-2) ppm. MS (ESI): *m/z* = 447.2 $[M]^+$, 403.4, 381.4, 325.3, 200.1. HRMS (ESI): *m/z* calcd. for $C_{24}H_{35}{}^{79}BrN_2O^+$: 447.2006, found: 447.2000 $[M]^+$; *m/z* calcd. for $C_{24}H_{35}{}^{81}BrN_2O^+$: 449.1988, found: 449.1957 $[M]^+$. DSC: Cr_1 49 °C [29.7 kJ/mol] Cr_2 57 °C [0.6 kJ/mol] SmA 63 °C [2.6 kJ/mol] I (2nd heating); I 65 °C [-3.6 kJ/mol] SmA 18 °C [-25.8 kJ/mol] Cr (2nd cooling).

2-[4-(4-Bromobutoxy)phenyl]-5-dodecylpyrimidine (*p*-17(12,4)).

From *p*-**16**(12) (0.70 mmol, 237 mg) and $Br(CH_2)_4Br$ (1.40 mmol, 301 mg); yield: 280 mg (84%), colourless solid. R_f = 0.35. Found: C, 65.63; H, 8.12; N, 5.73%. $C_{26}H_{39}BrN_2O$ requires C, 65.67; H, 8.27; N, 5.89%; M, 475.50. FT-IR (ATR): ν_{max}/cm^{-1} 2924 (s), 2853 (m), 1608 (m), 1585 (m), 1542 (w), 1514 (w), 1431 (s), 1252 (m), 1168 (m), 1043 (w), 909 (w), 846 (w), 798 (m), 734 (w), 654 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.20–1.40 (m, 16H, CH_2), 1.59–1.67 (m, 2H, CH_2), 1.94–2.01 (m, 2H, CH_2), 2.05–2.13 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5-CH_2), 3.50 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.07 (t, 2H, *J* = 5.9 Hz, OCH_2), 6.93–7.03 (m, 2H, 2'-H, 6'-H), 8.30–8.40 (m, 2H, 3'-H, 5'-H), 8.57 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 27.9, 29.0, 29.3, 29.45, 29.53, 29.61, 29.64, 30.1, 30.8, 31.9, 33.4 (CH_2), 66.9 (OCH_2), 114.4 (C-2', C-6'), 129.4 (C-3', C-5'), 130.4 (C-4'), 132.2 (C-5), 157.0 (C-4, C-6), 160.8 (C-1'), 162.4 (C-2) ppm. MS (ESI): *m/z* = 447.2 $[M]^+$, 403.4, 381.4, 325.3, 200.1. HRMS (ESI): *m/z* calcd. for $C_{26}H_{39}{}^{79}BrN_2O^+$: 475.2319, found: 475.2315 $[M]^+$; *m/z* calcd. for $C_{26}H_{39}{}^{81}BrN_2O^+$: 477.2302, found: 477.2291 $[M]^+$. DSC: Cr_1 20 °C [-8.5

kJ/mol] Cr_2 66 °C [51.2 kJ/mol] I (2nd heating); I 71 °C [-5.0 kJ/mol] SmA 22 °C [-26.0 kJ/mol] Cr (2nd cooling).

2-{4-[(8-Bromooctyl)oxy]phenyl}-5-octylpyrimidine (*p*-17(8,8)). From *p*-**16**(8) (0.35 mmol, 100 mg) and $Br(CH_2)_8Br$ (0.70 mmol, 190 mg); yield: 120 mg, (72 %). R_f = 0.41 (hexanes/ethyl acetate 20 : 1). Found: C, 64.90; H, 8.24; N, 5.73%. $C_{26}H_{39}BrN_2O$ requires C, 65.67; H, 8.27; N, 5.89%; M, 475.50. FT-IR (ATR): ν_{max}/cm^{-1} 2927 (s), 2855 (m), 2357 (w), 2023 (w), 1608 (w), 1584 (m), 1542 (w), 1515 (w), 1466 (w), 1433 (s), 1395 (w), 1329 (w), 1301 (w), 1256 (m), 1169 (w), 1108 (w), 1021 (w), 912 (w), 846 (w), 798 (m), 724 (w), 653 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.20–1.40 (m, 18H, CH_2), 1.60–1.68 (m, 2H, CH_2), 1.94–2.02 (m, 2H, CH_2), 2.05–2.13 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5-CH_2), 3.50 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.06 (t, 2H, *J* = 6.0 Hz, OCH_2), 6.94–7.00 (m, 2H, 2'-H, 6'-H), 8.32–8.38 (m, 2H, 3'-H, 5'-H), 8.57 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.6, 27.9, 29.0, 29.2, 29.3, 29.5, 30.2, 30.8, 31.8, 33.4 (CH_2), 66.9 (OCH_2), 114.4 (C-2', C-6'), 129.4 (C-3', C-5'), 130.4 (C-4'), 132.2 (C-5), 157.0 (C-4, C-6), 160.9 (C-1'), 162.4 (C-2) ppm. DSC: Cr 37 °C [34.6 kJ/mol] SmA 58 °C [4.4 kJ/mol] I (2nd heating); I 62 °C [-4.4 kJ/mol] SmA 19 °C [-25.3 kJ/mol] Cr (2nd cooling).

2-{4-[(8-Bromooctyl)oxy]phenyl}-5-decylpyrimidine (*p*-17(10,8)).[9] From *p*-**16**(10) (0.70 mmol, 220 mg) and $Br(CH_2)_8Br$ (1.40 mmol, 381 mg); yield: 251 mg (71%), colourless solid. R_f = 0.35 (hexanes/ethyl acetate 40 : 1). Found: C, 66.81; H, 8.46; N, 5.41%. $C_{28}H_{43}BrN_2O$ requires C, 66.78; H, 8.61; N, 5.41%; M, 503.56. FT-IR (ATR): ν_{max}/cm^{-1} 2925 (s), 2854 (m), 1608 (m), 1585 (m), 1542 (w), 1515 (w), 1466 (w), 1429 (s), 1394 (w), 1328 (w), 1301 (w), 1253 (m), 1168 (m), 1108 (w), 1022 (w), 908 (w), 846 (w), 798 (m), 731 (m), 653 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.21–1.53 (m, 22H, CH_2), 1.59–1.68 (m, 2H, CH_2), 1.77–1.91 (m, 4H, CH_2), 2.56–2.63 (m, 2H, 5-CH_2), 3.41 (t, 2H, *J* = 6.9 Hz, CH_2Br), 4.03 (t, 2H, *J* = 6.9 Hz, OCH_2), 6.94–7.01 (m, 2H, 2'-H, 6'-H), 8.32–8.38 (m, 2H, 3'-H, 5'-H), 8.57 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 26.0, 28.1, 28.7, 29.0, 29.2, 29.3, 29.4, 29.5, 29.6, 30.2, 30.8, 31.9, 32.8, 34.0 (CH_2), 68.0 (OCH_2), 114.4 (C-2', C-6'), 129.4 (C-3', C-5'), 130.2 (C-4'), 132.1 (C-5), 157.0 (C-4, C-6), 161.1 (C-1'), 162.5 (C-2) ppm. MS (ESI): *m/z* = 503.3 $[M]^+$, 463.3, 441.3, 423.3, 366.3, 301.1. HRMS (ESI): *m/z* calcd. for $C_{28}H_{43}{}^{79}BrN_2O^+$: 503.2632, found: 503.2640 $[M]^+$; *m/z* calcd. for $C_{28}H_{43}{}^{81}BrN_2O^+$: 505.2615, found: 505.2640 $[M]^+$. DSC: Cr 41 °C [32.7 kJ/mol] SmA 67 °C [3.1 kJ/mol] I (2nd heating); I 68 °C [-4.9 kJ/mol] SmA 35 °C [-35.3 kJ/mol] Cr (2nd cooling).

2-{4-[(8-Bromooctyl)oxy]phenyl}-5-dodecylpyrimidine (*p*-17(12,8)). From *p*-**16**(12) (1.18 mmol, 400 mg) and $Br(CH_2)_8Br$ (2.35 mmol, 639 mg); yield: 482 mg (77%), colourless solid. R_f = 0.48 (hexanes/ethyl acetate 20 : 1). Found: C, 67.72; H, 8.77; N, 5.16%. $C_{30}H_{47}BrN_2O$ requires C, 67.78; H, 8.91; N, 5.27%; M, 531.61. FT-IR (ATR): ν_{max}/cm^{-1} 2922 (s), 2852 (m), 1607 (m), 1584 (m), 1542 (w), 1514 (w), 1467 (w), 1429 (s), 1393 (w), 1328 (w), 1300 (w), 1250 (s), 1167 (m), 1107 (w), 1021 (w), 904 (w), 845 (w), 798 (m), 724 (s), 652 (m), 615 (w), 563 (w). δ_H (250 MHz, $CDCl_3$) 0.81 (t, 3H, *J* = 6.6 Hz, CH_3), 1.02–1.46 (m, 26H, CH_2), 1.49–1.64 (m, 2H, 5-CH_2CH_2), 1.66–1.90 (m, 4H, $OCH_2CH_2(CH_2)_4CH_2CH_2Br$), 2.41–2.61 (m, 2H, 5-$CH_2$), 3.34 (t, 2H, *J* = 6.9 Hz, CH_2Br), 3.95 (t, 2H, *J* = 6.5 Hz, OCH_2), 6.80–7.00 (m, 2H, 3'-H, 5'-H), 8.18–8.36 (m, 2H, 2'-H, 6'-H), 8.50 (s, 2H, 4-H, 6-H) ppm. δ_C (63 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 26.0, 28.1, 28.7, 29.0, 29.2, 29.4, 29.5, 29.6, 30.2, 30.8, 31.9, 32.8, 34.0 (CH_2), 68.0 (OCH_2), 114.4 (C-3', C-5'), 129.4 (C-2', C-6'), 130.2 (C-1'), 132.1 (C-5), 157.0 (C-4, C-6), 161.1 (C-4'), 162.5 (C-2) ppm. MS (ESI): *m/z* = 533.3 $[M]^+$. HRMS (ESI): *m/z* calcd. for $C_{30}H_{47}{}^{79}BrN_2O^+$: 531.2945, found: 531.2948 $[M]^+$; *m/z* calcd. for $C_{30}H_{47}{}^{81}BrN_2O^+$: 533.2929, found: 533.2915 $[M]^+$. DSC: Cr 58 °C [45.7 kJ/mol] SmA 71 °C [6.9 kJ/mol] I (2nd heating); I 73 °C [-8.1 kJ/mol] SmA 44 °C [-45.1 kJ/mol] Cr (2nd cooling).

2-[3-(4-Bromobutoxy)phenyl]-5-dodecylpyrimidine (*m*-17(12,4)). From *m*-**16**(12) (1.62 mmol, 550 mg) and $Br(CH_2)_4Br$ (3.23 mmol, 697 mg); yield: 661 mg (86%), colourless solid. R_f = 0.32, mp 51 °C. Found: C, 65.56; H, 8.16; N, 5.77%. $C_{26}H_{39}BrN_2O$ requires C, 65.67; H, 8.27; N, 5.89%; M, 475.50. FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2853 (s), 1723 (w), 1600 (w), 1586 (w), 1543 (m), 1445 (s), 1423 (s), 1391 (w), 1327 (m), 1288 (m), 1248 (m), 1207 (s), 1044 (w), 953 (w), 881 (w), 782 (m), 690 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.20–1.42 (m, 18H, CH_2), 1.62–1.70 (m, 4H, CH_2), 1.93–2.02 (m, 2H, CH_2), 2.06–2.14 (m, 2H, CH_2), 2.58–2.66 (m, 2H, 5-CH_2), 3.51 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.11 (t, 2H, *J* = 6.0 Hz, OCH_2), 6.98–7.03 (m, 1H, 6'-H), 7.38 (t, 1H, *J* = 7.9 Hz, 5'-H), 7.93–7.98 (m, 1H, 2'-H), 7.99–8.04 (m, 1H, 4'-H), 8.62 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 27.9, 29.4, 29.5, 29.63, 29.65, 30.2, 30.8, 31.9, 33.5 (CH_2), 66.9 (OCH_2), 113.0 (C-2'), 117.4 (C-6'), 120.5 (C-4'), 129.6 (C-5'), 133.2 (C-3'), 139.1 (C-5), 157.0 (C-4, C-6), 159.3 (C-1'), 162.4 (C-2) ppm. MS (ESI): *m/z* = 475.2 $[M+H]^+$, 395.3, 353.3, 341.3, 325.3, 199.1, 134.9. HRMS (ESI): *m/z* calcd. for $C_{26}H_{40}{}^{79}BrN_2O^+$: 475.2319; found: 475.2307 $[M+H]^+$; calcd. for $C_{26}H_{40}{}^{81}BrN_2O^+$: 477.2302; found: 477.2296 $[M+H]^+$.

2-{3-[(8-Bromooctyl)oxy]phenyl}-5-dodecylpyrimidine (*m*-17(12,8)). From *m*-**16**(12) (0.37 mmol, 126 mg) and $Br(CH_2)_8Br$ (0.74 mmol, 201 mg); yield: 136 mg (69%), colourless solid. R_f = 0.36. Found: C, 67.57; H, 8.82; N, 5.06%. $C_{30}H_{47}BrN_2O$ requires C, 67.78; H, 8.91; N,

5.27%; M, 531.61. FT-IR (ATR): ν_{max}/cm^{-1} 2925 (w), 2854 (w), 2253 (w), 1586 (w), 1544 (w), 1445 (w), 1424 (w), 1391 (w), 1327 (w), 1288 (w), 1235 (w), 1208 (w), 1090 (w), 1032 (w), 903 (s), 782 (w), 725 (s), 650 (m), 571 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.20–1.54 (m, 26H, CH_2), 1.61–1.69 (m, 4H, CH_2), 1.77–1.90 (m, 4H, CH_2), 2.58–2.66 (m, 2H, 5-CH_2), 3.41 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.07 (t, 2H, *J* = 6.6 Hz, OCH_2), 6.99–7.03 (m, 1H, 6'-H), 7.38 (t, 1H, *J* = 7.9 Hz, 5'-H), 7.94–7.98 (m, 1H, 2'-H), 7.98–8.02 (m, 1H, 4'-H), 8.62 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 26.0, 29.4, 28.1, 28.7, 29.04, 29.19, 29.27, 29.34, 29.53, 29.61, 29.64, 30.2, 30.8, 31.9, 32.8, 34.0 (CH_2), 68.0 (OCH_2), 113.0 (C-2'), 117.5 (C-6'), 120.2 (C-4'), 129.6 (C-5'), 133.1 (C-3'), 139.0 (C-5), 157.0 (C-4, C-6), 159.5 (C-1'), 162.5 (C-2) ppm. MS (ESI): *m/z* = 531.3 $[M]^+$. HRMS (ESI): *m/z* calcd. for $C_{30}H_{47}{}^{79}BrN_2O^+$: 531.2945; found: 531.2937 $[M]^+$; calcd. for $C_{30}H_{47}{}^{81}BrN_2O^+$: 533.2929; found: 531.2893 $[M]^+$. DSC: Cr 39 °C [47.0 kJ/mol] I (2nd heating); I 29 °C [-43.4 kJ/mol] Cr (2nd cooling).

Methyl 2-tetradecylpyrimidine-5-carboxylate (19). To a suspension of amidinium salt **10b** (1.00 g, 3.60 mmol) in abs. DMF (12 mL) in a dried flask was added **18** (830 mg, 4.18 mmol) and one drop of conc. HCl, and the reaction mixture was stirred at room temperature for 8 h. Then H_2O (20 mL) was added and the mixture extracted with CH_2Cl_2 (3 × 10 mL). The combined organic layers were dried (Na_2SO_4) and the solvent was removed under vacuum. The residue was purified by chromatography on SiO_2 (hexanes/EtOAc 20 : 1) to give **19** (675 mg, 56%) as a colorless solid. R_f = 0.22; mp 86 °C. Found: C, 72.18; H, 10.31; N, 8.13%. $C_{20}H_{34}N_2O_2$ requires C, 71.81; H, 10.25; N, 8.37%; M, 334.50. FT-IR (ATR): ν_{max}/cm^{-1} 2952 (m), 2912 (s), 2849 (s), 1720 (vs), 1594 (s), 1550 (m), 1472 (m), 1434 (s), 1302 (s), 1275 (m), 1126 (s), 1034 (m), 905 (s), 730 (s), 649 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.21–1.43 (m, 22H, CH_2), 1.78–1.89 (m, 2H, CH_2), 2.99–3.06 (m, 2H, 2-CH_2), 3.98 (s, 3H, CO_2CH_3), 9.20 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 28.6, 29.36, 29.41, 29.5, 29.61, 29.65, 29.68, 29.7, 31.9 (CH_2), 39.8 (2-CH_2), 52.5 (CO_2CH_3), 121.2 (C-5), 158.1 (C-4, C-6), 164.5 (CO_2CH_3), 175.4 (C-2) ppm. MS (ESI): *m/z* = 335.3 $[M]^+$, 293.2, 264.2, 241.3, 191.2. HRMS (ESI): *m/z* calcd. for $C_{20}H_{34}N_2O_2{}^+$: 335.2693, found: 335.2682 $[M]^+$.

Preparation of bromoalkyl 2-tetradecylpyrimidine-5-carboxylates 20(m). a) Following ref.[22] to a solution of **19** (230 mg, 0.67 mmol) in ethanol (10 mL) was added a 2 M aqueous NaOH solution (1.00 mL) and the reaction mixture stirred at room temperature for 2 h. Then the reaction mixture was neutralized with 2 N HCl, diluted with H_2O (10 mL) and extracted with Et_2O (3 × 15 mL). The combined organic layers were dried ($MgSO_4$) and concentrated to

give 2-tetradecylpyrimidine-5-carboxylic acid (213 mg, 99 %) as a colourless solid which was reacted without further purification. Mp 105 °C. Found: C, 71.11; H, 9.94; N, 8.72%. $C_{19}H_{32}N_2O_2$ requires C, 71.21; H, 10.06; N, 8.74%; M, 320.47. FT-IR (ATR): ν_{max}/cm^{-1} 2912 (s), 2848 (s), 1669 (m), 1594 (m), 1552 (w), 1471 (m), 1424 (s), 1303 (m), 904 (s), 725 (s), 649 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H J = 6.9 Hz, CH_3), 1.18–1.46 (m, 20H, CH_2), 1.79–1.94 (m, 4H, CH_2), 3.07–3.14 (m, 2H, 2-CH_2), 9.36 (s, 2H, 4-H, 6-H), 10.25 (s, br, 1H, CO_2H) ppm. δ_C NMR (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 28.6, 29.35, 29.38, 29.47, 49.49, 29.54, 29.62, 29.63, 29.65, 31.9 (CH_2), 39.4 (2-CH_2), 121.4 (C-5), 158.7 (C-4, C-6), 167.4 (CO_2H), 175.5 (C-2) ppm. MS (ESI): *m/z* = 320.2 $[M]^+$, 165.1, 151.1, 139.1. HRMS (ESI): *m/z* calcd. for $C_{19}H_{32}N_2O_2^+$: 320.2464, found: 320.2472 $[M]^+$.

b) To a solution of 2-tetradecylpyrimidine-5-carboxylic acid (1.00 mmol) in abs. CH_2Cl_2 (10 mL) was added successively *N*,*N*-dicyclohexylcarbodiimide (1.10 mmol) and 4-dimethylaminopyridine (3.50 mmol) and the reaction mixture was stirred at room temperature for 5 min. Then the respective ω-bromo-α-alkanol (1.10 mmol) was added and the reaction mixture stirred for a further 12 h. The reaction mixture was filtered and the filtrate concentrated. The crude product was purified by chromatography on SiO_2 (hexanes/EtOAc 1 : 1) to give the products as colourless solid.

4-Bromobutyl 2-tetradecylpyrimidine-5-carboxylate (20(4)). From 2-tetradecyl-pyrimidine-5-carboxylic acid (100 mg, 0.31 mmol) and $Br(CH_2)_4OH$ (57.0 mg, 0.37 mmol); yield: 100 mg (71%), mp 40 °C. Found: C, 60.93; H, 8.54; N, 6.11%. $C_{23}H_{39}BrN_2O_2$ requires C, 60.65; H, 8.63; N, 6.15%; M, 455.47. FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2853 (m), 1729 (s), 1590 (s), 1441 (m), 1390 (m), 1288 (s), 1137 (m), 1037 (w), 903 (s), 724 (s), 649 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.22–1.42 (m, 22H, CH_2), 1.78–1.88 (m, 2H, CH_2), 1.93–2.07 (m, 4H, CH_2), 2.98–3.08 (m, 2H, 2-CH_2), 3.48 (t, 2H, *J* = 6.6, CH_2Br), 4.41 (t, 2H, *J* = 6.0, OCH_2), 9.19 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 27.3, 28.6, 29.2, 29.36, 29.41, 29.5, 29.6, 29.65, 29.68, 29.69, 31.9 (CH_2), 32.8 (CH_2Br), 39.8 (2-CH_2), 64.6 (OCH_2), 121.3 (C-5), 158.1 (C-4, C-6), 163.9 (CO_2), 175.5 (C-2) ppm. MS (ESI): *m/z* = 455.2 $[M+H]^+$, 375.3, 321.3, 252.1, 133.1. HRMS (ESI): *m/z* calcd. for $C_{23}H_{40}{}^{79}BrN_2O_2^+$: 455.2268, found: 455.2270 $[M+H]^+$; calcd. for $C_{27}H_{40}{}^{81}BrN_4O_2^+$: 457.2250, found: 457.2261 $[M+H]^+$.

8-Bromooctyl 2-tetradecylpyrimidine-5-carboxylate (20(8)). From 2-tetradecylpyrimidine-5-carboxylic acid (220 mg, 0.69 mmol) and $Br(CH_2)_8OH$ (173 mg, 0.83 mmol); yield: 240 mg (61%), mp 34 °C. FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2853 (s), 1729 (s), 1590 (m), 1551 (m), 1441 (m), 1390 (w), 1287 (s), 1129 (s), 1037 (w), 908 (w), 724 (w), 647 (w). δ_H (500 MHz,

$CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.18–1.49 (m, 28H, CH_2), 1.70–1.89 (m, 6H, CH_2), 2.98–3.06 (m, 2H, 2-CH_2), 3.41 (t, 2H, *J* = 6.6 Hz, CH_2Br), 4.36 (t, 2H, *J* = 6.6 Hz, OCH_2), 9.19 (s, 2H, 4-H, 6-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.5, 25.8, 28.0, 28.57, 28.58, 28.63, 29.0, 29.36, 29.41, 29.5, 29.6, 29.65, 29.68, 29.69, 31.9, 32.7, 33.9 (CH_2), 39.8 (2-CH_2), 65.7 (OCH_2), 121.5 (C-5), 158.1 (C-4, C-6), 164.1 (CO_2), 175.3 (C-2) ppm. MS (ESI): *m/z* = 511.3 $[M+H]^+$, 431.4, 321.3, 225.2, 143.0. HRMS (ESI): *m/z* calcd. for $C_{27}H_{48}{}^{79}BrN_2O_2{}^+$: 511.2894, found: 511.2883 $[M+H]^+$; calcd. for $C_{27}H_{48}{}^{81}BrN_2O_2{}^+$: 513.2877, found: 513.2867 $[M+H]^+$.

General procedure for the preparation of alkylimidazolium bromides *p*-5(n,m), *p*-6(n,m), *m*-5(n,m), *m*-6(n,m) and 21(m). To a solution of the respective bromide *p*-**17**(n,m), *m*-**17**(12,m) or **20**(m) (1.00 mmol) in abs. acetonitrile was added freshly distilled *N*-methylimidazole **13a** or *N*-butylimidazole **13b** (10.0 mmol) and the reaction mixture stirred at 60 °C for the given time. Then the mixture was added dropwise in EtOAc (10 mL), the precipitate was filtered off and washed with EtOAc (3 × 10 mL). The precipitate was dried over P_4O_{10} to give the imidazolium salts as colourless solids.

3-Methyl-1-{4-[4-(5-octylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-5(8,4)). From *p*-**17**(8,4) (0.24 mmol, 100 mg) and **13a** (2.38 mmol, 196 mg) in abs. DMF (2 mL), reaction time 120 h; yield: 77.0 mg (65%), colourless solid. Found: C, 61.51; H, 7.30; N, 10.96%. $C_{26}H_{37}BrN_4O$ requires C, 62.27; H, 7.44; N, 11.17%; M, 501.50. FT-IR (ATR): ν_{max}/cm^{-1} 3093 (w), 2924 (m), 2854 (w), 1606 (m), 1582 (m), 1542 (w), 1514 (w), 1428 (s), 1398 (w), 1328 (w), 1300 (w), 1253 (m), 1168 (m), 1108 (w), 1018 (w), 904 (s), 846 (w), 799 (m), 724 (s), 652 (m), 623 (w). δ_H (250 MHz, $CDCl_3$) 0.80 (t, 3H, *J* = 6.6 Hz, CH_3), 1.09–1.40 (m, 10H, CH_2), 1.44–1.69 (m, 2H, NCH_2CH_2), 1.71–1.95 (m, 2H, 5'''-CH_2CH_2), 2.01–2.24 (m, 2H, OCH_2CH_2), 2.45–2.65 (m, 2H, 5'''-CH_2), 3.92–4.15 (m, 5H, NCH_3, OCH_2), 4.33–4.52 (m, 2H, NCH_2), 6.83–6.97 (m, 2H, 3''-H, 5''-H), 7.21–7.44 (m, 2H, 4-H, 5-H), 8.21–8.33 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.51 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.6, 25.9, 27.4, 29.1, 29.2, 29.3, 30.2, 30.8, 31.8 (CH_3) 36.7 (NCH_3), 49.8 (CH_2N), 67.0 (OCH_2), 114.4 (C-3'', C-5''), 122.0 (C-4), 123.2 (C-5), 129.5 (C-2'', C-6''), 130.7 (C-1''), 132.3 (C-2), 137.9 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-4''), 162.2 (C-2''') ppm. MS (ESI): *m/z* = 421.3 $[M-Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{26}H_{37}N_4O^+$: 421.2962, found: 421.2963 $[M-Br]^+$. DSC: Cr_1 11 °C [1.7 kJ/mol] Cr_2 74 °C [6.0 kJ/mol] Cr_3 99 °C [9.6 kJ/mol] SmA 190 °C [0.8 kJ/mol] I (2nd heating); I 190 °C [-0.8 kJ/mol] SmA 53 °C [-13.5 kJ/mol] Cr_2 7 °C [-0.9 kJ/mol] Cr_2 (2nd cooling).

3-Methyl-1-{4-[4-(5-decylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide

(*p*-5(10,4)). From *p*-**17**(10,4) (0.096 mmol, 43.0 mg) and **13a** (0.96 mmol, 79.0 mg) in abs. DMF (2 mL), reaction time 120 h; yield: 20.0 mg (39%), colourless solid. Found: C, 61.94; H, 7.84; N, 9.59%. $C_{28}H_{41}BrN_4O$ requires C, 63.51; H, 7.80; N, 10.58%; M, 529.56. FT-IR (ATR): ν_{max}/cm^{-1} 3150 (w), 3078 (w), 2956 (w), 2922 (s), 2852 (m), 1738 (w), 1606 (m), 1582 (m), 1542 (w), 1514 (w), 1467 (w), 1429 (s), 1397 (w), 1327 (w), 1299 (w), 1252 (s), 1165 (m), 1109 (w), 1066 (w), 1034 (w), 1018 (w), 935 (w), 847 (w), 798 (m), 767 (w), 722 (w), 654 (w), 616 (w), 587 (w), 560 (w). δ_H (250 MHz, $CDCl_3$) 0.80 (t, 3H, *J* = 6.6 Hz, CH_3), 1.09–1.38 (m, 14H, CH_2), 1.48–1.65 (m, 2H, NCH_2CH_2), 1.78–1.90 (m, 2H, 5'''-CH_2CH_2), 2.05–2.12 (m, 2H, OCH_2CH_2), 2.46–2.60 (m, 2H, 5'''-CH_2), 3.94–4.09 (m, 5H, NCH_3, OCH_2), 4.41 (t, 2H, *J* = 7.3 Hz, NCH_2), 6.84–6.94 (m, 2H, 3''-H, 5''-H), 7.21–7.34 (m, 2H, 4-H, 5-H), 8.20–8.33 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.63 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 27.4, 29.1, 29.3, 29.5, 29.6, 30.2, 30.8, 31.9 (CH_2) 36.8 (NCH_3), 49.9 (CH_2N), 67.0 (OCH_2), 114.4 (C-3'', C-5''), 121.8 (C-4), 123.0 (C-5), 129.5 (C-2'', C-6''), 130.7 (C-1''), 132.3 (C-2), 138.3 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-4''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 449.3 $[M\text{-}Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{28}H_{41}N_4O^+$: 449.3275, found: 449.3285 $[M\text{-}Br]^+$. DSC: Cr_1 63 °C [22.2 kJ/mol] Cr_2 69 °C [6.4 kJ/mol] SmA 220 °C [0.5 kJ/mol] I (1st heating, decomposition upon clearing).

3-Methyl-1-{4-[4-(5-dodecylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-5(12,4)). From *p*-**17**(12,4) (0.21 mmol, 100 mg) and **13a** (2.10 mmol, 172 mg) in abs. DMF (2 mL), reaction time 120 h; yield: 84.0 mg (72%), colourless solid. Found: C, 64.81; H, 8.05; N, 9.86%. $C_{30}H_{45}BrN_4O$ requires C, 64.62; H, 8.13; N, 10.05%; M, 557.61. FT-IR (ATR): ν_{max}/cm^{-1} 2922 (m), 2852 (w), 2194 (w), 1607 (w), 1584 (m), 1541 (w), 1514 (w), 1468 (w), 1428 (s), 1397 (w), 1328 (w), 1301 (w), 1251 (m), 1167 (m), 1109 (w), 1037 (w), 1020 (w), 908 (m), 846 (w), 798 (m), 727 (s), 653 (w), 642 (w), 619 (w), 555 (w), 534 (w). δ_H (250 MHz, $CDCl_3$ 0.80 (t, 3H, *J* = 6.6 Hz, CH_3), 1.07–1.39 (m, 18H, CH_2), 1.47–1.66 (m, 2H, NCH_2CH_2), 1.79–1.92 (m, 2H, 5'''-CH_2CH_2), 2.03–2.21 (m, 2H, OCH_2CH_2), 2.46–2.60 (m, 2H, 5'''-CH_2), 3.97–4.08 (m, 5H, NCH_3, OCH_2), 4.41 (t, 2H, *J* = 7.3 Hz, NCH_2), 6.82-6.95 (m, 2H, 3''-H, 5''-H), 7.24–7.37 (m, 2H, 4-H, 5-H), 8.20–8.33 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.54 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 27.4, 29.1, 29.3, 29.5, 29.6, 30.2, 30.8, 31.9 (CH_2) 36.8 (NCH_3), 49.9 (CH_2N), 67.0 (OCH_2), 114.4 (C-3'', C-5''), 121.9 (C-4), 123.1 (C-5), 129.5 (C-2'', C-6''), 130.7 (C-1''), 132.3 (C-2), 138.0 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-4''), 162.2 (C-2''') ppm. MS (ESI): *m/z* = 477.4 $[M\text{-}Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{30}H_{45}N_4O^+$: 477.3588, found: 477.3592 $[M\text{-}Br]^+$. DSC: Cr_1 71 °C [32.8 kJ/mol] Cr_2 112 °C [21.5 kJ/mol] SmA 255 °C [0.7

kJ/mol] I (1st heating, decomposition upon clearing).

3-Methyl-1-{4-[4-(5-octylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*p*-5(8,8)). From *p*-**17**(8,8) (0.11 mmol, 50.0 mg) and **13a** (1.05 mmol, 86.0 mg) in abs. DMF (2 mL), reaction time 120 h; yield: 28.0 mg (48%), colourless solid. Found: C, 63.79; H, 7.99; N, 9.53%. $C_{30}H_{45}BrN_4O$ requires C, 64.62; H, 8.13; N, 10.05%; M, 557.61. FT-IR (ATR): ν_{max}/cm^{-1} 2927 (m), 2855 (m), 2201 (w), 1608 (w), 1585 (m), 1542 (w), 1514 (w), 1466 (w), 1430 (s), 1393 (w), 1328 (w), 1300 (w), 1252 (m), 1168 (m), 1108 (w), 1023 (w), 907 (m), 846 (w), 798 (w), 729 (s), 653 (w), 623 (w). δ_H (250 MHz, $CDCl_3$) 0.81 (t, 3H, *J* = 6.6 Hz, CH_3), 1.10–1.48 (m, 18H, CH_2), 1.50–1.65 (m, 2H, NCH_2CH_2), 1.66–1.78 (m, 2H, 5'''-CH_2CH_2), 1.80–1.94 (m, 2H, OCH_2CH_2), 2.53 (t, 2H, *J* = 7.6 Hz, 5'''-CH_2), 3.95 (t, 2H, *J* = 6.4 Hz, OCH_2), 4.05 (s, 3H, NCH_3), 4.25 (t, 2H, *J* = 7.5 Hz, CH_2N), 6.86–6.97 (m, 2H, 3''-H, 5''-H), 7.13–7.17 (m, 1H, 4-H), 7.18–7.22 (m, 1H, 5-H overlap with $CHCl_3$), 8.21–8.34 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.66 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.1, 28.8, 29.0, 29.1, 29.2, 29.3, 30.2, 30.8, 31.8 (CH_2) 36.8 (NCH_3), 50.2 (CH_2N), 67.9 (OCH_2), 114.4 (C-3'', C-5''), 121.4 (C-4), 123.0 (C-5), 129.4 (C-2'', C-6''), 130.2 (C-1''), 132.2 (C-2), 138.4 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-4''), 162.4 (C-2''') ppm. MS (ESI): *m/z* = 477.4 $[M\text{-}Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{30}H_{45}N_4O^+$: 477.3588, found: 477.3590 $[M\text{-}Br]^+$. DSC: Cr_1 51 °C [5.1 kJ/mol] Cr_2 61 °C [2.4 kJ/mol] Cr_3 88 °C [18.7 kJ/mol] SmA 148 °C [0.9 kJ/mol] I (2nd heating); I 51 °C [-0.9 kJ/mol] SmA 48 °C [-31.6 kJ/mol] Cr (2nd cooling).

3-Methyl-1-{4-[4-(5-decylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*p*-5(10,8)). From *p*-**17**(10,8) (0.13 mmol, 64.0 mg) and **13a** (1.27 mmol, 104 mg) in abs. DMF (2 mL), reaction time 120 h; yield: 25.0 mg (34%), colourless solid. Found: C, 63.59; H, 8.57; N, 8.91%. $C_{32}H_{49}BrN_4O$ requires C, 65.63; H, 8.43; N, 9.57%; M, 585.66. FT-IR (ATR): ν_{max}/cm^{-1} 3141 (w), 3036 (w), 2921 (s), 2851 (m), 1735 (w), 1608 (m), 1584 (m), 1542 (w), 1514 (w), 1468 (w), 1431 (s), 1395 (w), 1329 (w), 1301 (w), 1249 (m), 1167 (m), 1107 (w), 1065 (w), 1036 (w), 904 (w), 844 (w), 817 (w), 797 (m), 723 (m), 653 (w), 622 (w), 564 (w), 549 (w). δ_H (250 MHz, $CDCl_3$) 0.81 (t, 3H, *J* = 6.7 Hz, CH_3), 1.09–1.48 (m, 22H, CH_2), 1.49–1.62 (m, 2H, NCH_2CH_2), 1.68–1.79 (m, 2H, 5'''-CH_2CH_2), 1.80–1.93 (m, 2H, OCH_2CH_2), 2.46–2.63 (m, 2H, 5'''-CH_2), 3.95 (t, 2H, *J* = 7.5 Hz, OCH_2), 4.05 (s, 3H, NCH_3), 4.25 (t, 2H, *J* = 7.5 Hz, CH_2N), 6.83–7.00 (m, 2H, 3''-H, 5''-H), 7.14–7.24 (m, 2H, 4-H, 5-H), 8.21–8.33 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.64 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.1, 28.8, 29.0, 29.1, 29.3, 29.5, 29.6, 30.2, 30.8, 31.9 (CH_2) 36.8 (NCH_3), 50.2 (CH_2N), 67.9 (OCH_2), 114.4 (C-3'', C-5''), 121.4 (C-4), 123.0 (C-5),

129.4 (C-2'', C-6''), 130.2 (C-1''), 132.1 (C-2), 138.3 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-4''), 162.4 (C-2''') ppm. MS (ESI): *m/z* = 505.4 $[M\text{-}Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{32}H_{49}N_4O^+$: 505.3901, found: 505.3906 $[M\text{-}Br]^+$. DSC: Cr 71 °C [25.1 kJ/mol] SmC 98 °C [0.4 kJ/mol] SmA 177 °C [0.7 kJ/mol] I (2nd heating); I 178 °C [-0.7 kJ/mol] SmA 102 °C [-0.5 kJ/mol] SmC 49 °C [-35.4 kJ/mol] Cr (2nd cooling).

3-Butyl-1-{4-[4-(5-octylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-6(8,4)). From *p*-**17**(8,4) (0.17 mmol, 70.0 mg) and **13b** (1.40 mmol, 174 mg) in MeCN (4 mL), reaction time 120 h; yield: 24.0 mg (26%), colourless solid. Found: C, 63.58; H, 8.27; N, 9.31%. $C_{29}H_{43}BrN_4O$ requires C, 64.08; H, 7.97; N, 10.31%; M, 543.58. FT-IR (ATR): ν_{max}/cm^{-1} 3070 (w), 2957 (w), 2927 (m), 2856 (w), 2361 (w), 1607 (m), 1585 (m), 1563 (w), 1542 (w), 1512 (w), 1464 (w), 1428 (s), 1397 (w), 1379 (w), 1328 (w), 1301 (w), 1251 (m), 1168 (m), 1109 (w), 1078 (w), 1029 (w), 965 (w), 916 (w), 848 (w), 800 (m), 751 (w), 665 (w), 654 (w). δ_H (250 MHz, $CDCl_3$) 0.81 (t, 3H, *J* = 6.6 Hz, CH_3), 0.89 [t, 3H, *J* = 7.3 Hz, $N(CH_2)_3CH_3$], 1.09–1.40 (m, 12H, CH_2), 1.49–1.64 (m, 2H, CH_2), 1.74–1.93 (m, 4H, CH_2), 2.02–2.23 (m, 2H, CH_2), 2.46–2.61 (m, 2H, 5'''-CH_2), 4.03 (t, 2H, *J* = 5.8 Hz, OCH_2), 4.25 [t, 2H, *J* = 7.4 Hz, $NCH_2(CH_2)_2CH_3$], 4.45 (t, 2H, *J* = 7.3 Hz, NCH_2) 6.83–6.95 (m, 2H, 3''-H, 5''-H), 7.18–7.35 (m, 2H, 4-H, 5-H overlap with $CHCl_3$), 8.19–8.34 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.73 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) δ = 13.4 $[N(CH_2)_3CH_3]$, 14.1 (CH_3), 19.5, 22.6, 25.9, 27.4, 29.0, 29.2, 29.3, 29.7, 30.2, 30.8, 31.8, 32.1 (CH_2), 49.8 (NCH_2), 50.0 (NCH_2), 67.0 (OCH_2), 114.4 (C-3'', C-5''), 121.5 (C-4), 121.8 (C-5), 129.5 (C-2'', C-6''), 130.7 (C-1''), 132.3 (C-2), 137.8 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-4''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 463.3 $[M\text{-}Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{29}H_{43}N_4O^+$: 463.3431, found: 463.3424 $[M\text{-}Br]^+$. DSC: Cr 56 °C [13.2 kJ/mol] I (1st heating).

3-Butyl-1-{4-[4-(5-decylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-6(10,4)). From *p*-**17**(10,4) (0.28 mmol, 125 mg) and **13b** (1.40 mmol, 174 mg) in MeCN (4 mL), reaction time 96 h; yield: 143 mg (89%). Found: C, 64.65; H, 8.10; N, 9.45%. $C_{31}H_{47}BrN_4O$ requires C, 65.13; H, 8.29; N, 9.80%; M, 571.64. FT-IR (ATR): ν_{max}/cm^{-1} 2925 (s), 2854 (m), 1607 (m), 1585 (m), 1562 (w), 1542 (w), 1514 (w), 1466 (m), 1429 (s), 1328 (m), 1252 (s), 1167 (m), 906 (s), 848 (w), 799 (m), 727 (s), 652 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 0.9 (t, 3H *J* = 7. Hz, CH_3), 1.20–1.42 (m, 16H, CH_2), 1.59–1.68 (m, 2H, CH_2), 1.85–1.94 (m, 4H, CH_2), 2.14–2.24 (m, 2H, CH_2), 2.55–2.63 (m, 2H, 5'''-CH_2), 4.09 (t, 2H, *J* = 5.7 Hz, OCH_2), 4.32 (t, 2H, *J* = 7.6 Hz, NCH_2), 4.52 (t, 2H, *J* = 7.3 Hz, NCH_2), 6.91–6.99 (m, 2H, 2''-H, 6''-H), 7.32–7.49 (m, 2H, 4-H, 5-H), 8.29–8.37 (m, 2H, 3''-H, 5''-H), 8.57 (s,

2H, 4'''-H, 6'''-H), 10.71 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 13.5, 14.1 (CH_3), 19.5, 22.7, 25.9, 27.4, 29.1, 29.29, 29.34, 29.5, 29.56, 30.2, 30.8, 31.9, 32.1 (CH_2), 49.8, 49.9 (NCH_2, CH_2N), 67.0 (OCH_2), 114.4 (C-2'', C-6''), 121.7, 122.0 (C-4, C-5), 129.5 (C-3'', C-5''), 130.6 (C-4''), 132.3 (C-2), 137.5 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-1''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 491.4 $[M\text{-}Br]^+$, 367.3, 325.2, 295.2, 199.1, 179.2, 151.1, 125.1. HRMS (ESI): *m/z* calcd. for $C_{31}H_{47}N_4O^+$: 491.3744, found: 491.3762 $[M\text{-}Br]^+$. DSC: Cr 97 °C [15.1 kJ/mol] I (2nd heating), I 69 °C [-16.9 kJ/mol] Cr (2nd cooling).

3-Butyl-1-{4-[4-(5-dodecylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-6(12,4)). From *p*-**17**(12,4) (0.21 mmol, 100.0 mg) and **13b** (2.10 mmol, 261 mg) in MeCN (4 mL), reaction time 120 h; yield: 79.0 mg (63%), colourless solid. Found: C, 66.12; H, 8.57; N, 8.86%. $C_{33}H_{51}BrN_4O$ requires C, 66.09; H, 8.57; N, 9.34%; M, 599.69. FT-IR (ATR): ν_{max}/cm^{-1} 3035 (w), 2956 (w), 2919 (m), 2873 (w), 2850 (m), 2363 (w), 2342 (w), 1606 (m), 1585 (m), 1565 (w), 1541 (w), 1514 (w), 1469 (w), 1431 (s), 1398 (w), 1328 (w), 1303 (w), 1255 (m), 1237 (w), 1166 (m), 1109 (w), 1067 (w), 1017 (w), 962 (w), 930 (w), 850 (w), 799 (m), 749 (w), 720 (w), 703 (w). δ_H (250 MHz, $CDCl_3$) 0.80 (t, 3H, *J* = 6.6 Hz, CH_3), 0.89 [t, 3H, *J* = 7.3 Hz, $N(CH_2)_3CH_3$], 1.08–1.40 (m, 20H, CH_2), 1.47–1.64 (m, 2H, CH_2), 1.74–1.91 (m, 4H, CH_2), 2.06–2.22 (m, 2H, CH_2), 2.45–2.59 (m, 2H, 5-CH_2), 4.03 (t, 2H, *J* = 5.8 Hz, OCH_2), 4.25 [t, 2H, *J* = 7.4 Hz, $NCH_2(CH_2)_2CH_3$], 4.45 (t, 2H, *J* = 7.5 Hz, NCH_2) 6.83–6.94 (m, 2H, 3''-H, 5''-H), 7.21–7.36 (m, 2H, 4-H, 5-H overlap with $CHCl_3$), 8.21–8.32 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.71 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 13.5 [$N(CH_2)_3CH_3$], 14.1 (CH_3), 19.5, 22.7, 25.9, 27.4, 29.1, 29.3, 29.5, 29.6, 30.2, 30.8, 31.9, 32.1 (CH_2), 49.8 (NCH_2), 50.0 (NCH_2) 67.0 (OCH_2), 114.3 (C-3'', C-5''), 121.5 (C-4), 121.8 (C-5), 129.5 (C-2'', C-6''), 130.7 (C-1''), 132.3 (C-2), 137.7 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-4''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 519.4 $[M\text{-}Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{33}H_{51}BrN_4O^+$: 519.1457, found: 519.4059 $[M\text{-}Br]^+$. DSC: Cr_1 35 °C [21.9 kJ mol^{-1}] Cr_2 46 °C [2.7 kJ mol^{-1}] SmA 113 °C [0.4 kJ mol^{-1}] I (2nd heating); I 113 °C [-0.4 kJ mol^{-1}] SmA 34 °C [-20.0 kJ mol^{-1}] Cr (2nd cooling).

3-Butyl-1-{8-[4-(5-octylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*p*-6(8,8)). From *p*-**17**(8,8) (0.15 mmol, 71.0 mg) and **13b** (1.49 mmol, 185 mg), 120 h; yield: 52.0 mg (58%). FT-IR (ATR): ν_{max}/cm^{-1} 3132 (w), 3067 (w), 2956 (m), 2924 (m), 2854 (m), 1733 (w), 1606 (m), 1584 (m), 1563 (w), 1542 (w), 1514 (w), 1464 (w), 1428 (s), 1398 (w), 1378 (w), 1328 (w), 1302 (w), 1251 (s), 1166 (s), 1109 (w), 1067 (w), 1020 (w), 965 (w), 938 (w), 849 (w), 800 (m). δ_H (250 MHz, $CDCl_3$) 0.81 (t, 3H, *J* = 6.6 Hz, CH_3), 0.90 [t, 3H, *J* = 7.3 Hz, $N(CH_2)_3CH_3$], 1.13–1.48 (m, 20H, CH_2), 1.49–1.64 (m, 2H, CH_2), 1.66–1.78 (m, 2H, CH_2),

1.79–1.95 (m, 4H, CH_2), 2.46–2.60 (m, 2H, 5'''-CH_2), 3.95 (t, 2H, J = 6.5 Hz, OCH_2), 4.29 [t, 4H, J = 7.4 Hz, NCH_2, $NCH_2(CH_2)_2CH_3$], 6.83–6.98 (m, 2H, 3''-H, 5''-H), 7.20–7.27 (m, 2H, 4-H, 5-H overlap with $CHCl_3$), 8.20–8.33 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.71 (s, 1H, 2-H) ppm. δ_C (63 MHz, $CDCl_3$) 13.5 [$N(CH_2)_3CH_3$], 14.1 (CH_2), 19,5, 22.6, 25.9, 26.1, 28.9, 29.0, 29.1, 29.2, 29.3, 30.2, 30,3, 30.8, 31.8, 32.2 (CH_2), 49.9 (NCH_2), 50.1 (NCH_2) 67.9 (OCH_2), 114.4 (C-3'', C-5''), 121.5 (C-4, C-5), 129.4 (C-2'', C-6''), 130.2 (C-1''), 132.2 (C-2), 137.8 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-4''), 162.4 (C-2''') ppm. MS (ESI): m/z = 519.4 $[M–Br]^+$. HRMS (ESI): m/z calcd. for $C_{35}H_{51}N_4O^+$: 519.4057, found: 519.4062 $[M–Br]^+$. DSC: Cr 82 °C [3.7 kJ/mol] I (2nd heating), I 87 °C [-4.9 kJ/mol] Cr (2nd cooling).

3-Butyl-1-{8-[4-(5-decylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*p*-6(10,8)). From *p*-**17**(10,8) (0.16 mmol, 80.0 mg) and **13b** (1.60 mmol, 197 mg), 96 h; yield: 87.0 mg (87%). FT-IR (ATR): ν_{max}/cm^{-1} 2925 (s), 2854 (m), 1608 (m), 1585 (m), 1562 (w), 1542 (w), 1514 (w), 1466 (m), 1432 (s), 1328 (m), 1252 (s), 1167 (m), 903 (s), 846 (w), 798 (m), 724 (s), 650 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, J = 6.9 Hz, CH_3), 0.97 (t, 3H J = 7.3 Hz, CH_3), 1.20–1.51 (m, 22H, CH_2), 1.60–1.68 (m, 2H, CH_2), 1.76–1.83 (m, 2H, CH_2), 1.88–1.98 (m, 4H, CH_2), 2.56–2.63 (m, 2H, 5'''-CH_2), 4.02 (t, 2H, J = 6.6 Hz, OCH_2), 4.36 (t, 2H, J = 7.3 Hz, NCH_2, CH_2N), 6.95–6.99 (m, 2H, 2''-H, 6''-H), 7.23–7.26 (m, 2H, 4-H, 5-H), 8.31–8.37 (m, 2H, 3''-H, 5''-H), 8.57 (s, 2H, 4'''-H, 6'''-H), 10.81 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 13.5, 14.1 (CH_3), 19.5, 22.7, 25.9, 26.1, 28.9, 29.02, 29.04, 29.1, 29.3, 29.4, 29.5, 29.6, 30.2, 30.3, 30.8, 30.9, 31.9, 32.2 (CH_2), 50.0 (NCH_2), 50.2 (CH_2N), 67.9 (OCH_2), 114.3 (C-2'', C-6''), 121.39, 121.42 (C-4, C-5), 129.4 (C-3'', C-5''), 130.2 (C-4''), 132.2 (C-2), 137.9 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-1''), 162.4 (C-2''') ppm. MS (ESI): m/z = 547.4 $[M–Br]^+$, 491.4, 423.3, 313.2, 295.2, 235.2, 207.2, 193.2, 179.2, 138.1, 125.1, 97.1. HRMS (ESI): m/z calcd. for $C_{35}H_{55}N_4O^+$: 547.4370, found: 547.4380 $[M–Br]^+$. DSC: Cr_1 22 °C [9.4 kJ/mol] Cr_2 32 °C [-16.2 kJ/mol] Cr_3 54 °C [21.7 kJ/mol] SmA 87 °C [6.1 kJ/mol] I (2nd heating), I 92 °C [-6.4 kJ/mol] SmA 26 °C [-9.3 kJ/mol] Cr (2nd cooling).

3-Butyl-1-{8-[4-(5-octylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*p*-6(12,8)). From *p*-**17**(12,8) (0.19 mmol, 100 mg) and **13b** (1.88 mmol, 234 mg), 120 h; yield: 91.0 mg (74%), colourless solid. FT-IR (ATR): ν_{max}/cm^{-1} 3131 (w), 3034 (w), 2954 (w), 2920 (s), 2849 (m), 1608 (m), 1584 (m), 1565 (w), 1541 (w), 1514 (w), 1469 (w), 1432 (s), 1396 (w), 1357 (w), 1330 (w), 1306 (w), 1252 (m), 1167 (m), 1107 (w), 1065 (w), 1036 (w), 1001 (w), 946 (w), 923 (w), 845 (w), 799 (m), 773 (w), 724 (w). δ_H (250 MHz, $CDCl_3$) 0.80 (t, 3H, J = 6.6 Hz, CH_3), 0.90 [t, 3H, J = 7.4 Hz, $N(CH_2)_3CH_3$], 1.07–1.47 (m, 28H, CH_2), 1.48–1.64 (m,

2H, CH_2), 1.65–1.78 (m, 2H, CH_2), 1.79–1.98 (m, 4H, CH_2), 2.45-2.61 (m, 2H, 5’’’-CH_2), 3.94 (t, 2H, *J* = 6.4 Hz, OCH_2), 4.29 [t, 4H, *J* = 7.4 Hz, NCH_2 , $NCH_2(CH_2)_2CH_3$], 6.85–6.96 (m, 2H, 3’’-H, 5’’-H), 7.21–7.28 (m, 2H, 4-H, 5-H), 8.21–8.34 (m, 2H, 2’’-H, 6’’-H), 8.50 (s, 2H, 4’’’-H, 6’’’-H), 10.70 (s, 1H, 2-H) ppm. δ_C (63 MHz, $CDCl_3$) 13.5 [$N(CH_2)_3CH_3$], 14.1 (CH_3), 19,5, 22.7, 25.9, 26.1, 28.9, 29.0, 29.1, 29.3, 29.5, 29.6, 30.2, 30,3, 30.8, 31.9, 32.2 (CH_2), 49.9 (NCH_2), 50.1 (NCH_2) 67.9 (OCH_2), 114.4 (C-3’’, C-5’’), 121.6 (C-4, C-5), 129.4 (C-2’’, C-6’’), 130.2 (C-1’’), 132.2 (C-2), 137.8 (C-5’’’), 157.0 (C-4’’’, C-6’’’), 161.1 (C-4’’), 162.4 (C-2’’’) ppm. MS (ESI): *m/z* = 575.5 $[M–Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{37}H_{59}N_4O^+$: 575.4683, found: 575.4693 $[M–Br]^+$. DSC: Cr_1 63 °C [21.7 kJ/mol] Cr_2 69 °C [4.0 kJ/mol] SmA 106 °C [1.0 kJ/mol] I (2nd heating), I 106 °C [-0.9 kJ mol^{-1}] SmA 72 °C [-3.7 kJ mol^{-1}] C 44 °C [-29.8 kJ mol^{-1}] Cr (2nd cooling).

1-{4-[3-(5-Dodecylpyrimidin-2-yl)phenoxy]butyl}-3-methylimidazolium bromide (*m*-5(12,4)). From *m*-**17**(12,4) (0.168 mmol, 80.0 mg) and **13a** (1.68 mmol, 138 mg), 72 h; yield: 71.0 mg (76%). Found: C, 62.46; H, 7.98; N, 9.73%. $C_{30}H_{45}BrN_4O_2$ requires C, 62.82; H, 7.91; N, 9.77%, M, 573.61. FT-IR (ATR): ν_{max}/cm^{-1} 3081 (m), 2923 (s), 2853 (s), 1585 (m), 1544 (m), 1446 (m), 1424 (s), 1392 (w), 1327 (m), 1290 (m), 1208 (m), 1167 (m), 903 (s), 781 (m), 725 (s). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.21–1.41 (m, 18H, CH_2), 1.61–1.70 (m, 2H, CH_2), 1.86–1.94 (m, 2H, CH_2), 2.14–2.42 (m, 2H, CH_2), 2.58–2.65 (m, 2H, 5’’’-CH_2), 4.09 (s, 3H, NCH_3), 4.12 (t, 2H, *J* = 5.7 Hz, OCH_2), 4.48 (t, 2H, *J* = 7.6 Hz, CH_2N), 6.97–7.01 (m, 1H, 6’’-H), 7.35–7.43 (m, 2H, 4-H, 5-H), 7.38 (t, 1H, *J* = 7.9, 5’’-H), 7.89-7.92 (s, 1H, 2’’-H), 7.99–8.03 (m, 1H, 4’’-H), 8.61 (s, 2H, 4’’’-H, 6’’’-H), 10.57 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 27.5, 29.1, 29.3, 29.5, 29.61, 29.64, 30.2, 30.8, 31.9 (CH_2), 36.8 (NCH_3), 49.9 (CH_2N), 67.1 (OCH_2), 113.2 (C-2’’), 117.0 (C-6’’), 120.7 (C-4’’), 121.9, 123.2 (C-4, C-5), 129.8 (C-5’’), 133.3 (C-2), 138.0 (C-3’’), 139.2 (C-5’’’), 157.0 (C-4’’’, C-6’’’), 158.9 (C-1’’), 162.2 (C-2’’’) ppm. MS (ESI): *m/z* = 477.4 $[M–Br]^+$, 395.3, 367.3, 353.3, 341.3, 325.3, 199.1, 137.1. HRMS (ESI): *m/z* calcd. for $C_{30}H_{45}N_4O_2^+$: 477.3588, found: 477.3586 $[M–Br]^+$. DSC: Cr 81 °C [3.4 kJ/mol] SmA 185 °C [0.6 kJ/mol] I (2nd heating), I 184 °C [-0.7 kJ/mol] SmA (2nd cooling); no recrystallization was observed upon cooling.

3-Butyl-1-{4-[3-(5-dodecylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*m*-6(12,4)). From *m*-**17**(12,4) (0.168 mmol, 80.0 mg) and **13b** (1.68 mmol, 209 mg), 96 h; yield: 83.0 mg (82%). FT-IR (ATR): ν_{max}/cm^{-1} 3068 (m), 2956 (m), 2924 (s), 2854 (s), 1586 (m), 1544 (m), 1446 (m), 1425 (s), 1392 (w), 1327 (m), 1290 (m), 1208 (m), 1164 (m), 1038 (w), 782 (m), 654 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 0.97 (t, 3H, *J* =

7.3 Hz, CH_3), 1.21–1.43 (m, 20H, CH_2), 1.61–1.69 (m, 2H, CH_2), 1.86–1.96 (m, 4H, CH_2), 2.16–2.25 (m, 2H, CH_2), 2.59–2.65 (m, 2H, 5'''-CH_2), 4.13 (t, 2H, J = 5.7 Hz, OCH_2), 4.33 (t, 2H, J = 7.6 Hz, NCH_2), 4.53 (t, 2H, J = 7.3 Hz, CH_2N), 6.97–7.01 (m, 1H, 6''-H), 7.23–7.36 (m, 2H, 4-H, 5-H), 7.38 (t, 1H, J = 7.9, 5''-H), 7.89–7.94 (s, 1H, 2''-H), 7.98–8.05 (m, 1H, 4''-H), 8.63 (s, 2H, 4'''-H, 6'''-H), 10.85 (s, 1H, 2-H) ppm. δ_C NMR (125 MHz, $CDCl_3$) 13.5, 14.1 (CH_3), 19.5, 22.7, 25.9, 27.5, 29.1, 29.3, 29.53, 29.61, 29.64, 30.2, 30.8, 31.9, 32.1 (CH_2), 49.8, 50.0 (CH_2N, NCH_2), 67.1 (OCH_2), 113.3 (C-2''), 117.0 (C-6''), 120.7 (C-4''), 121.4, 121.7 (C-4, C-5), 129.8 (C-5''), 133.3 (C-2), 137.9 (C-3''), 139.2 (C-5'''), 157.0 (C-4''', C-6'''), 158.9 (C-1''), 162.2 (C-2''') ppm. MS (ESI): *m/z* = 519.4 $[M–Br]^+$, 395.3, 367.3, 353.3, 325.3, 179.2, 125.1. HRMS (ESI): *m/z* calcd. for $C_{33}H_{51}N_4O^+$: 519.4057, found: 519.4068 $[M–Br]^+$. DSC: Cr_1 24 °C [-8.3 kJ/mol] Cr_2 38 °C [7.1 kJ/mol] I (2nd heating), I 19 °C [-0.1 kJ/mol] Cr (2nd cooling).

3-Butyl-1-{4-[3-(5-dodecylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*m*-6(12,8)). From *m*-**17**(12,8) (0.15 mmol, 80.0 mg) and **13b** (1.50 mmol, 186 mg), 96 h; yield: 82.0 mg (84%). FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2852 (s), 1585 (m), 1544 (m), 1446 (m), 1425 (s), 1328 (m), 1290 (m), 1208 (m), 1165 (m), 1038 (w), 904 (s), 728 (m), 651 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, J = 6.9 Hz, CH_3), 0.97 (t, 3H J = 7.3 Hz, CH_3), 1.21–1.54 (m, 26H, CH_2), 1.61–1.69 (m, 2H, CH_2), 1.76–1.85 (m, 2H, CH_2), 1.88–1.97 (m, 4H, CH_2), 2.59–2.66 (m, 2H, 5'''-CH_2), 4.06 (t, 2H, J = 6.3 Hz, OCH_2), 4.36 (t, 2 x 2H, J = 7.6 Hz, NCH_2, CH_2N), 6.99–7.03 (m, 1H, 6''-H), 7.23–7.26 (m, 2H, 4-H, 5-H), 7.38 (t, 1H, J = 7.9, 5''-H), 7.93–7.96 (s, 1H, 2'''-H), 7.98–8.02 (m, 1H, 4''-H), 8.63 (s, 2H, 4'''-H, 6'''-H), 10.85 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 13.5, 14.1 (CH_3), 19.5, 22.7, 25.9, 26.1, 26.3, 28.8, 28.9, 29.1, 29.2, 29.3, 29.53, 29.61, 29.64, 30.19, 30.24, 30.8, 31.9, 32.2 (CH_2), 49.9, 50.2 (CH_2N, NCH_2), 67.9 (OCH_2), 113.0 (C-2''), 117.3 (C-6''), 120.2 (C-4''), 121.38, 121.41 (C-4, C-5), 129.6 (C-5''), 133.1 (C-2), 137.8 (C-3''), 139.1 (C-5'''), 157.0 (C-4''', C-6'''), 159.5 (C-1''), 162.4 (C-2''') ppm. MS (ESI): *m/z* = 575.5 $[M-Br]^+$, 519.4, 451.4, 341.3, 235.2, 207.2, 193.2, 179.2, 125.1. HRMS (ESI): *m/z* calcd. for $C_{37}H_{59}N_4O^+$: 575.4683, found: 575.4681 $[M-Br]^+$. DSC: Cr 40 °C [33.3 J/mol] SmA 89 °C [2.2 kJ/mol] I (2nd heating); I 81 °C [-4.5 kJ/mol] SmA 40 °C [-36.3 kJ/mol] Cr (2nd cooling).

3-Methyl-1-(4-{[(2-tetradecylpyrimidin-5-yl)carbonyl]oxy}butyl)imidazolium bromide (21(4)). From **20**(4) (0.13 mmol, 60.0 mg) and **13a** (1.30 mmol, 107 mg), 72 h; yield: 53.0 mg (77%). Found: C, 58.59; H, 8.49; N, 10.00%. $C_{27}H_{45}BrN_4O_2$ requires C, 60.32; H, 8.44; N, 10.42%; M, 537.58. FT-IR (ATR): ν_{max}/cm^{-1} 2919 (s), 2851 (m), 1721 (s), 1590 (s), 1443 (m), 1390 (m), 1292 (s), 1168 (m), 1138 (m), 1039 (w), 906 (s), 730 (s), 648 (m).

δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, J = 6.9 Hz, CH_3), 1.22–1.42 (m, 22H, CH_2), 1.78–1.87 (m, 2H, CH_2), 1.88–1.96 (m, 2H, CH_2), 2.08–2.16 (m, 2H, CH_2), 2.99–3.05 (m, 2H, 2-CH_2), 4.11 (s, 3H, NCH_3), 4.44 (t, 2H, J = 6.6 Hz, OCH_2), 4.52 (t, 2H, J = 7.3 Hz, CH_2N), 7.29–7.37 (m, 2H, 4-H, 5-H), 9.16 (s, 2H, 4''-H, 6''-H), 10.80 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.6, 27.1, 28.6, 29.36, 29.39, 29.42, 29.5, 29.6, 29.65, 29.68, 31.9 (CH_2), 36.8 (NCH_3), 39.8 (2''-CH_2), 49.6 (CH_2N), 64.3 (OCH_2), 121.1 (C-5''), 121.7, 123.0 (C-4, C-5), 138.6 (C-2), 158.1 (C-4'', C-6''), 164.0 (CO_2), 175.6 (C-2'') ppm. MS (ESI): *m/z* = 457.4 $[M–Br]^+$, 375.3, 321.3, 303.2, 233.2, 163.1, 137.1. HRMS (ESI): *m/z* calcd. for $C_{27}H_{45}N_4O_2^+$: 457.3537, found: 457.3531 $[M–Br]^+$. DSC: Cr 44 °C [35.3 kJ/mol] SmA 237 °C [0.5 kJ/mol] I (1st heating); decomposition upon clearing.

General procedure for the preparation of dodecyl- and dodecyloxyphenylimidazolium bromides *p*-7(n,m), *p*-8(n,m), *m*-7(n,m), *m*-8(n,m), 22(m) and 23(m). To a solution of the respective bromide (1.00 mmol) in acetonitrile (6 mL) was added *N*-dodecylimidazole **13c** (10.0 mmol) or 1-(4-dodecyloxyphenyl)imidazole **13d** (3.00 mmol), and the reaction mixture was stirred at 60 °C for the given time. Then the mixture was added dropwise in Et_2O (10 mL), the precipitate was filtered off and washed with Et_2O (3 × 10 mL). The precipitate was dried over P_4O_{10} under vacuum to give the imidazolium salts as colourless solids.

3-Dodecyl-1-{4-[4-(5-octylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-7(8,4)). From *p*-**17**(8,4) (0.27 mmol, 110 mg) and **13c** (2.70 mmol, 634 mg), 96 h; yield: 160 mg (91%). Found: C, 67.77; H, 9.01; N, 8.44%. $C_{37}H_{59}BrN_4O$ requires C, 67.76; H, 9.07; N, 8.54%; M, 655.79. FT-IR (ATR): ν_{max}/cm^{-1} 2925 (s), 2854 (m), 1607 (m), 1585 (m), 1515 (w), 1466 (m), 1430 (s), 1328 (m), 1252 (s), 1168 (m), 905 (s), 849 (w), 799 (m), 728 (s), 650 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, J = 6.9 Hz, CH_3), 0.88 (t, 3H J = 6.9 Hz, CH_3), 1.18–1.40 (m, 28H, CH_2), 1.59–1.68 (m, 2H, CH_2), 1.85–1.96 (m, 4H, CH_2), 2.15–2.25 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5'''-CH_2), 4.09 (t, 2H, J = 5.7 Hz, OCH_2), 4.30 (t, 2H, J = 7.3 Hz, NCH_2), 4.53 (t, 2H, J = 7.3 Hz, CH_2N), 6.92–6.97 (m, 2H, 2''-H, 6''-H), 7.24–7.42 (m, 2H, 4-H, 5-H), 8.31–8.37 (m, 2H, 3''-H, 5''-H), 8.57 (s, 2H, 4'''-H, 6'''-H), 10.78 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.08, 14.12 (CH_3), 22.6, 22.7, 25.9, 26.3, 27.4, 29.0, 29.1, 29.2, 29.31, 29.32, 29.4, 29.5, 29.6, 30.2, 30.3, 30.8, 31.1, 31.8, 31.9 (CH_2), 49.8 (NCH_2), 50.2 (CH_2N), 67.0 (OCH_2), 114.3 (C-2'', C-6''), 121.5, 121.8 (C-4, C-5), 129.5 (C-3'', C-5''), 130.7 (C-4''), 132.3 (C-2), 137.8 (C-5'''), 156.8 (C-4''', C-6'''), 160.5 (C-1''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 575.5 $[M–Br]^+$, 407.3, 339.2, 297.2, 269.2, 237.2. HRMS (ESI): *m/z* calcd. for $C_{37}H_{59}N_4O^+$: 575.4683, found: 575.4677 $[M–Br]^+$. DSC: Cr_1 85 °C [1.3

kJ/mol] Cr_2 110 °C [23.9 kJ/mol] I (2nd heating), I 100 °C [-1.8 kJ/mol] SmA 93 °C [-27.0 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-{4-[4-(5-decylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-7(10,4)). From *p*-**17**(10,4) (0.28 mmol, 125 mg) and **13c** (1.40 mmol, 330 mg), 96 h; yield: 175 mg (91 %). Found: C, 68.63; H, 9.24; N, 7.89%. $C_{39}H_{63}BrN_4O$ requires C, 68.50; H, 9.29; N, 8.19%; M, 683.85. FT-IR (ATR): ν_{max}/cm^{-1} 3063 (w), 2956 (m), 2920 (s), 2851 (s), 1606 (m), 1586 (m), 1566 (w), 1541 (w), 1514 (m), 1467 (m), 1431 (s), 1329 (m), 1304 (w), 1258 (s), 1166 (s), 1109 (m), 1033 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 0.88 (t, 3H *J* = 6.9 Hz, CH_3), 1.19–1.41 (m, 34H, CH_2), 1.59–1.67 (m, 2H, CH_2), 1.85–1.95 (m, 4H, CH_2), 2.15–2.24 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5’’’-CH_2), 4.09 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.30 (t, 2H, *J* = 7.6 Hz, NCH_2), 4.53 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.92–6.98 (m, 2H, 2’’-H, 6’’-H), 7.25–7.43 (m, 2H, 4-H, 5-H), 8.28–8.38 (m, 2H, 3’’-H, 5’’-H), 8.57 (s, 2H, 4’’’-H, 6’’’-H), 10.76 (s, 1H, 2-H) ppm. δ_C NMR (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.3, 27.4, 29.0, 29.30, 29.33, 29.35, 29.37, 29.5, 29.53, 29.56, 29.6, 30.2, 30.3, 30.8, 31.88, 31.89 (CH_2), 49.8 (NCH_2), 50.2 (CH_2N), 67.0 (OCH_2), 114.3 (C-2’’, C-6’’), 121.5, 121.9 (C-4, C-5), 129.5 (C-3’’, C-5’’), 130.7 (C-4’’), 132.3 (C-2), 137.3 (C-5’’’), 156.9 (C-4’’’, C-6’’’), 160.5 (C-1’’), 162.3 (C-2’’’) ppm. MS (ESI): *m/z* = 603.5 $[M–Br]^+$, 435.3, 367.3, 339.2, 325.2, 313.2, 297.2. HRMS (ESI): *m/z* calcd. for $C_{39}H_{63}N_4O^+$: 603.4996, found: 603.4984 $[M–Br]^+$. DSC: Cr_1 4 °C [4.2 kJ/mol] Cr_2 112 °C [28.7 kJ/mol] I (2nd heating), I 116 °C [-1.8 kJ/mol] SmA 106 °C [-30.4 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-{4-[4-(5-dodecylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide (*p*-7(12,4)). From *p*-**17**(12,4) (0.13 mmol, 62.0 mg) and **13c** (1.30 mmol, 308 mg), in DMF, 120 h; yield: 61 mg (66 %). Found: C, 69.02; H, 9.45; N, 7.66%. $C_{41}H_{67}BrN_4O$ requires C, 69.17; H, 9.49; N, 7.87%; M, 711.9. FT-IR (ATR): ν_{max}/cm^{-1} 3064 (w), 2955 (w), 2920 (s), 2851 (m), 1722 (w), 1607 (m), 1588 (m), 1563 (w), 1542 (w), 1514 (w), 1468 (m), 1433 (s), 1394 (w), 1329 (w), 1303 (w), 1253 (m), 1168 (m), 1111 (w), 1067 (w), 1035 (w), 964 (w), 908 (w), 845 (w), 796 (m), 724 (m), 654 (w), 640 (w), 617 (w). δ_H (250 MHz, $CDCl_3$) 0.74–0.89 (m, 6H, CH_3, $NCH_2(CH_2)_{11}CH_3$), 1.05–1.37 (m, 36H, CH_2), 1.48–1.64 (m, 2H, CH_2), 1.71–1.93 (m, 4H, CH_2), 2.02–2.22 (m, 2H, CH_2), 2.46–2.61 (m, 2H, 5’’’-CH_2), 4.03 (t, 2H, *J* = 5.9 Hz, OCH_2), 4.23 (t, 2H, *J* = 7.6 Hz, $NCH_2(CH_2)_{11}CH_3$), 4.46 (t, 2H, *J* = 7.2 Hz, CH_2N), 6.83–6.94 (m, 2H, 3’’-H, 5’’-H), 7.17–7.36 (m, 2H, 4-H, 5-H), 8.21–8.34 (m, 2H, 2’’-H, 6’’-H), 8.50 (s, 2H, 4’’’-H, 6’’’-H), 10.73 (s, 1H, 2-H) ppm. δ_C NMR (63 MHz, $CDCl_3$) 14.1 (CH_3, $NCH_2(CH_2)_{11}CH_3$), 22.7, 25.8, 26.3, 27.4, 29.0, 29.1, 29.3, 29.5, 29.6, 30.2, 30.3, 30.8, 31.9 (CH_2), 49.8 (NCH_2), 50.2 (CH_2N), 67.0 (OCH_2), 114.3 (C-3’’, C-5’’), 121.5 (C-4), 121.8

(C-5), 129.5 (C-2'', C-6''), 130.7 (C-1''), 132.3 (C-2), 137.8 (C-5'''), 157.0 (C-4''', C-6'''), 160.5 (C-4''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 631.5 $[M–Br]^+$, 463.3, 395.3, 367.3, 353.3, 341.3, 323.2, 291.3. HRMS (ESI): *m/z* calcd. for $C_{41}H_{67}N_4O^+$: 631.5309, found: 631.5302 $[M–Br]^+$. DSC: Cr_1 23 °C [4.8 kJ/mol] Cr_2 111 °C [30.9 kJ/mol] I (2nd heating), I 125 °C [-1.6 kJ/mol] SmA 94 °C [-32.3 kJ/mol] Cr_2 21 °C [-4.4 kJ/mol] Cr_1 (2nd cooling).

3-Dodecyl-1-{8-[4-(5-octylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide ((*p*-7(8,8)). From *p*-**17**(8,8) (0.15 mmol, 70.0 mg) and **13c** (1.50 mmol, 348 mg), 96 h; yield: 70.0 mg (67%). Found: C, 68.50; H, 9.34; N, 7.55%. $C_{41}H_{67}BrN_4O$ requires C, 69.17; H, 9.49; N, 7.87%; M, 711.90. FT-IR (ATR): ν_{max}/cm^{-1} 3034 (w), 2955 (m), 2921 (s), 2851 (s), 1608 (m), 1585 (m), 1563 (w), 1542 (w), 1515 (m), 1467 (m), 1431 (s), 1330 (m), 1302 (w), 1253 (s), 1166 (m), 1108 (m), 1037 (m), 908 (w), 844 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 7.0 Hz, $2CH_3$), 0.88 (t, 3H *J* = 7.0 Hz, $2CH_3$), 1.19–1.51 (m, 34H, CH_2), 1.59–1.67 (m, 2H, CH_2), 1.76–1.83 (m, 2H, CH_2), 1.87–1.97 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5'''-CH_2), 4.02 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.34 (t, 2H, *J* = 7.3 Hz, NCH_2), 4.37 (t, 2H, *J* = 7.6 Hz, CH_2N), 6.94–6.99 (m, 2H, 2''-H, 6''-H), 7.22–7.26 (m, 2H, 4-H, 5-H), 8.29–8.37 (m, 2H, 3''-H, 5''-H), 8.57 (s, 2H, 4'''-H, 6'''-H), 10.80 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.09, 14.13 (2 x CH_3), 22.6, 22.7, 25.9, 26.1, 26.3, 28.9, 29.0, 29.04, 29.13, 29.2, 29.3, 29.33, 29.4, 29.5, 29.6, 30.2, 30.3, 30.3, 30.8, 31.8, 31.9 (CH_2), 50.15 (NCH_2), 50.21 (CH_2N), 67.9 (OCH_2), 114.4 (C-2'', C-6''), 121.40, 121.43 (C-4, C-5), 129.4 (C-3'', C-5''), 130.2 (C-4''), 132.2 (C-2), 137.9 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-1''), 162.4 (C-2''') ppm. MS (ESI): *m/z* = 631.5 $[M–Br]^+$, 463.3, 395.3, 347.3, 319.3, 305.3, 285.2. HRMS (ESI): *m/z* calcd. for $C_{41}H_{67}N_4O^+$: 631.5309, found: 631.5310 $[M–Br]^+$. DSC: Cr 74 °C [42.1 kJ/mol] SmA 79 °C [1.7 kJ/mol] I (2nd heating), I 79 °C [-4.2 kJ/mol] SmA 58 °C [-43.0 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-{8-[4-(5-decylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide ((*p*-7(10,8)). From *p*-**17**(8,8) (0.16 mmol, 80.0 mg) and **13c** (1.60 mmol, 376 mg), 96 h; yield: 98.0 mg (83%). FT-IR (ATR): ν_{max}/cm^{-1} 2954 (m), 2921 (s), 2851 (s), 1609 (m), 1585 (m), 1562 (w), 1542 (w), 1514 (m), 1468 (m), 1433 (s), 1331 (m), 1304 (w), 1254 (s), 1166 (m), 1107 (m), 1037 (m), 844 (w), 798 (m) cm. δ_H (500 MHz, $CDCl_3$) 0.87 (t, 6H, *J* = 6.9 Hz, 2 x CH_3), 1.19–1.42 (m, 40H, CH_2), 1.43–1.52 (m, 2H, CH_2), 1.59–1.69 (m, 2H, CH_2), 1.75–1.84 (m, 2H, CH_2), 1.87–1.99 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 5'''-CH_2), 4.02 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.34 (t, 2H, *J* = 7.3 Hz, NCH_2), 4.37 (t, 2H, *J* = 7.6 Hz, CH_2N), 6.94–7.00 (m, 2H, 2''-H, 6''-H), 7.20–7.25 (m, 2H, 4-H, 5-H), 8.30–8.37 (m, 2H, 3''-H, 5''-H), 8.57 (s, 2H, 4'''-H, 6'''-H), 10.86 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.1,

26.3, 28.9, 29.03, 29.04, 29.13, 29.3, 29.33, 29.35, 29.4, 29.5, 29.53, 29.57, 29.6, 30.2, 30.3, 30.8, 31.88, 31.90 (CH_2), 50.16 (NCH_2), 50.22 (CH_2N), 67.9 (OCH_2), 114.4 (C-2'', C-6''), 121.3, 121.4 (C-4, C-5), 129.4 (C-3'', C-5''), 130.2 (C-4''), 132.1 (C-2), 138.0 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-1''), 162.5 (C-2''') ppm. MS (ESI): *m/z* = 659.6 $[M–Br]^+$, 491.4, 423.3, 347.3, 305.3, 277.3, 237.2, 207.2, 179.2, 137.1. HRMS (ESI): *m/z* ber. für $C_{43}H_{71}N_4O_2^+$: 659.5622, gef.: 659.5616 für $[M–Br]^+$. DSC: Cr 78 °C [54.9 kJ/mol] I (2nd heating), I 82 °C [-3.6 kJ/mol] SmA 65 °C [-50.7 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-{4-[4-(5-dodecylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*p*-7(12,8)). From *p*-**17**(12,8) (0.11 mmol, 60.0 mg) and **13c** (1.13 mmol, 277 mg), in DMF, 120 h; yield: 52 mg (60 %). Found: C, 68.64; H, 9.81; N, 7.00%. $C_{45}H_{75}BrN_4O$ requires C, 70.37; H, 9.84; N, 7.30%; M, 768.01. FT-IR (ATR): ν_{max}/cm^{-1} 2920 (s), 2851 (m), 1608 (w), 1585 (m), 1563 (w), 1542 (w), 1514 (w), 1468 (w), 1431 (s), 1396 (w), 1330 (w), 1253 (m), 1167 (m), 1108 (w), 1037 (w), 905 (m), 845 (w), 798 (m), 725 (s), 651 (m). δ_H (250 MHz, $CDCl_3$) 0.80 (t, 6H, *J* = 6.6 Hz, CH_3, $NCH_2(CH_2)_{11}CH_3$), 1.08–1.46 (m, 44H, CH_2), 1.48–1.64 (m, 2H, CH_2), 1.65–1.77 (m, 2H, CH_2), 1.78–1.95 (m, 4H, CH_2), 2.46–2.60 (m, 2H, 5'''-CH_2), 3.95 (t, 2H, *J* = 6.5 Hz, OCH_2), 4.22–4.39 (m, 4H, CH_2N, $NCH_2(CH_2)_{11}CH_3$), 6.84–6.97 (m, 2H, 3''-H, 5''-H), 7.13–7.20 (m, 2H, 4-H, 5-H), 8.22–8.33 (m, 2H, 2''-H, 6''-H), 8.50 (s, 2H, 4'''-H, 6'''-H), 10.75 (s, 1H, 2-H) ppm. δ_C NMR (63 MHz, $CDCl_3$) 14.1 (CH_3, $NCH_2(CH_2)_{11}CH_3$), 22.7, 25.9, 26.1, 26.3, 38.9, 29.0, 29.1, 29.3, 29.5, 29.6, 30.2, 30.3, 30.8, 31.9 (CH_2), 50.2 (NCH_2, CH_2N), 67.9 (OCH_2), 114.4 (C-3'', C-5''), 121.4 (C-4, C-5), 129.4 (C-2'', C-6''), 130.2 (C-1''), 132.1 (C-2), 137.9 (C-5'''), 157.0 (C-4''', C-6'''), 161.1 (C-4''), 162.3 (C-2''') ppm. MS (ESI): *m/z* = 687.6 $[M–Br]^+$, 545.4, 519.4, 451.4, 423.3, 353.3, 341.3, 319.3, 305.3, 291.3, 277.3. HRMS (ESI): *m/z* calcd. for $C_{45}H_{75}N_4O^+$: 687.5935, found: 687.5916 $[M–Br]^+$. DSC: Cr 81 °C [53.2 kJ/mol] I (2nd heating), I 81 °C [-2.4 kJ/mol] Cr_2 71 °C [-56.1 kJ/mol] Cr_1 (2nd cooling).

1-{4-[4-[5-Octylpyrimidin-2-yl)phenoxy]butyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide ((*p*-8(8,4)). From *p*-**17**(8,4) (0.27 mmol, 110 mg) and **13d** (0.80 mmol, 264 mg), 72 h; yield: 177 mg (88%). Found: C, 68.93; H, 8.46; N, 7.42%. $C_{43}H_{63}BrN_4O_2$ requires C, 69.06; H, 8.49; N, 7.49%; M, 747.89. FT-IR (ATR): ν_{max}/cm^{-1} 2922 (s), 2853 (m), 1607 (m), 1586 (m), 1551 (w), 1512 (m), 1468 (m), 1429 (s), 1304 (m), 1253 (s), 1168 (m), 1072 (m), 904 (s), 725 (s), 650 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 6H, *J* = 6.9 Hz, 2 x CH_3), 1.19–1.49 (m, 26H, CH_2), 1.59–1.68 (m, 2H, CH_2), 1.74–1.82 (m, 2H, CH_2), 1.91–1.99 (m, 2H, CH_2), 2.20–2.31 (m, 2H, CH_2), 2.55–2.62 (m, 2H, 2'''-CH_2), 3.96 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.10 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.72 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.91–6.97 (m, 2H, 2''-H, 6''-H),

6.97–7.04 (m, 2H, 3’’’’-H, 5’’’’-H), 7.49–7.61 (m, 2H, 4-H, 5-H), 7.61–7.67 (m, 2H, 2’’’’-H, 6’’’’-H), 8.28–8.35 (m, 2H, 3’’-H, 5’’-H), 8.56 (s, 2H, 4’’’-H, 6’’’-H), 11.00 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.08, 14.13 (CH_3), 22.64, 22.69, 25.9, 26.0, 27.4, 29.1, 29.2, 29.3, 29.35, 29.38, 29.57, 29.61, 29.64, 29.7, 30.2, 30.8, 31.8, 31.9 (CH_2), 50.0 (NCH_2), 67.0, 68.7 (OCH_2), 114.4 (C-2’’, C-6’’), 116.1 (C-3’’’’, C-5’’’’), 120.5, 122.5 (C-4, C-5), 123.3 (C-2’’’’, C-6’’’’), 127.1 (C-1’’’’), 129.5 (C-3’’, C-5’’), 130.6 (C-4’’), 132.3 (C-5’’’), 136.1 (C-2), 157.0 (C-4’’’, C-6’’’), 160.45, 160.55 (C-1’’, C-4’’’’), 162.3 (C-2’’’) ppm. MS (ESI): *m/z* = 667.5 $[M–Br]^+$, 499.3, 383.3, 355.3, 339.2, 311.2, 297.2. HRMS (ESI): *m/z* calcd. for $C_{43}H_{63}N_4O_2^+$: 667.4946, found: 667.4940 $[M–Br]^+$. DSC: Cr 158 °C [35.2 kJ/mol] SmA 190 °C [2.4 kJ/mol] I (2nd heating), I 189 °C [-2.2 kJ/mol] SmA 140 °C [-35.9 kJ/mol] Cr (2nd cooling).

1-{4-[4-[5-Decylpyrimidin-2-yl)phenoxy]butyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide ((*p*-8(10,4)). From *p*-**17**(10,4) (0.28 mmol, 125 mg) and **13d** (0.56 mmol, 184 mg), 72 h; yield: 209 mg (96%). Found: C, 69.43; H, 8.52; N, 7.02%. $C_{45}H_{67}BrN_4O_2$ requires C, 69.65; H, 8.70; N, 7.22%; M, 775.94. FT-IR (ATR): ν_{max}/cm^{-1} 3063 (w), 2922 (s), 2852 (m), 1607 (m), 1585 (m), 1547 (m), 1514 (s), 1468 (m), 1429 (vs), 1311 (w), 1255 (s), 1171 (m), 1068 (m), 1021 (m), 904 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 0.88 (t, 3H *J* = 6.9 Hz, CH_3), 1.20–1.50 (m, 32H, CH_2), 1.59–1.68 (m, 2H, CH_2), 1.75–1.84 (m, 2H, CH_2), 1.91–2.03 (m, 2H, CH_2), 2.21–2.32 (m, 2H, CH_2), 2.56–2.65 (m, 2H, 2’’’-CH_2), 3.97 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.11 (t, 2H, *J* = 5.7 Hz, OCH_2), 4.73 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.92–6.97 (m, 2H, 2’’-H, 6’’-H), 6.99–7.04 (m, 2H, 3’’’’-H, 5’’’’-H), 7.46–7.53 (m, 2H, 4-H, 5-H), 7.61–7.66 (m, 2H, 2’’’’-H, 6’’’’-H), 8.31–8.35 (m, 2H, 3’’-H, 5’’-H), 8.57 (s, 2H, 4’’’-H, 6’’’-H), 11.09 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.67, 22.69, 25.9, 26.0, 27.5, 29.0, 29.3, 29.35, 29.37, 29.5, 29.57, 29.60, 29.64, 29.7, 30.2, 30.8, 31.88, 31.92 (CH_2), 50.1 (NCH_2), 67.0, 68.7 (2 x OCH_2), 114.4 (C-2’’, C-6’’), 116.1 (C-3’’’’, C-5’’’’), 120.4, 122.3 (C-4, C-5), 123.3 (C-2’’’’, C-6’’’’), 127.1 (C-1’’’’), 129.5 (C-3’’, C-5’’), 130.6 (C-4’’), 132.3 (C-5’’’), 136.1 (C-2), 157.0 (C-4’’’, C-6’’’), 160.48, 160.53 (C-1’’, C-4’’’’), 162.3 (C-2’’’) ppm. MS (ESI): *m/z* = 695.5 $[M–Br]^+$, 383.3, 367.3, 355.3, 325.2, 313.22, 295.2. HRMS (ESI): *m/z* calcd. for $C_{45}H_{67}N_4O_2^+$: 695.5259, found: 695.5262 $[M–Br]^+$. DSC: Cr 156 °C [38.7 kJ/mol] SmA 204 °C [2.3 kJ/mol] I (1st heating), I 187 °C [-2.0 kJ/mol] SmA 122 °C [-25.9 kJ/mol] Cr (1st cooling).

1-{4-[4-[5-Dodecylpyrimidin-2-yl)phenoxy]butyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide ((*p*-8(12,4)). From *p*-**17**(12,4) (0.11 mmol, 50.0 mg) and **13d** (1.05 mmol, 345 mg), in DMF, 120 h; yield: 57.0 mg (68%). Found: C, 70.18; H, 8.78; N, 6.65%. $C_{47}H_{71}BrN_4O_2$

requires C, 70.21; H, 8.90; N, 6.97%; M, 613.71. FT-IR (ATR): ν_{max}/cm^{-1} 3058 (w), 2920 (s), 2852 (m), 1607 (m), 1586 (m), 1550 (m), 1514 (m), 1468 (w), 1431 (s), 1395 (w), 1328 (w), 1256 (s), 1169 (m), 1110 (w), 1070 (w), 1021 (w), 958 (w), 904 (m), 851 (w), 831 (w), 800 (m), 722 (m), 653 (w), 618 (w). δ_H (250 MHz, $CDCl_3$) 0.75–0.84 (m, 6H, CH_3), 1.10–1.31 (m, 34H, CH_2), 1.34–1.42 (m, 2H, CH_2), 1.50–1.61 (m, 2H, 5’’’-CH_2CH_2), 1.66–1.77 (m, 2H, OCH_2CH_2), 1.84–1.94 (m, 2H, OCH_2CH_2), 2.12–2.27 (m, 2H, NCH_2), 2.52 (t, 2H, *J* = 7.9 Hz, 5’’’-CH_2), 3.89 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.03 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.65 (t, 2H, *J* = 7.3 Hz, NCH_2), 6.84–6.90 (m, 2H, 3’’-H, 5’’-H), 6.91–6.96 (m, 2H, 3’’’’-H, 5’’’’-H), 7.42–7.51 (m, 2H, 4-H, 5-H), 7.54–7.59 (m, 2H, 2’’’’-H, 6’’’’-H), 8.23–8.27 (m, 2H, 2’’-H, 6’’-H), 8.49 (s, 2H, 4’’’-H, 6’’’-H), 10.94 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.0, 27.5, 29.1, 29.4, 29.5, 29.6, 29.7, 30.2, 30.8, 31.9 (CH_2), 50.1 (NCH_2), 67.0, 68.7 (2 x OCH_2), 114.4 (C-3’’, C-5’’), 116.1 (C-3’’’’, C-5’’’’), 120.5, 122.4 (C-4, C-5), 123.3 (C-2’’’’, C-6’’’’), 127.1 (C-1’’’’, C-5’’’, C-1’’), 129.5 (C-2’’, C-6’’), 130.6, 132.3 (C-1’’’’, C-5’’’, C-1’’), 136.2 (C-2), 157.0 (C-4’’’, C-6’’’), 160.5, 160.6 (C-4’’, C-4’’’’), 162.3 (C-2’’’) ppm. MS (ESI): *m/z* = 723.6 $[M–Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{47}H_{71}N_4O_2^+$: 723.5572, found: 723.5580 $[M–Br]^+$. DSC: Cr 143 °C [34.2 kJ/mol] SmA 195 °C [1.9 kJ/mol] I (2nd heating), I 196 °C [-2.0 kJ/mol] SmA 128 °C [-34.5 kJ/mol] Cr (2nd cooling).

1-{8-[4-[5-Octylpyrimidin-2-yl)phenoxy]octyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide ((*p*-8(8,8)). From *p*-**17**(8,8) (0.19 mmol, 90.0 mg) and **13d** (0.57 mmol, 187 mg), 72 h; yield: 144 mg (95%). Found: C, 69.94; H, 8.85; N, 6.88%. $C_{47}H_{71}BrN_4O_2$ requires C, 70.21; H, 8.90; N, 6.97%; M, 803.99. FT-IR (ATR): ν_{max}/cm^{-1} 2922 (s), 2854 (m), 1607 (m), 1584 (m), 1568 (w), 1551 (w), 1514 (m), 1468 (m), 1428 (s), 1302 (m), 1255 (s), 1166 (m), 1075 (m), 903 (s), 723 (s), 650 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 6H, *J* = 6.9 Hz, 2 x CH_3), 1.21–1.51 (m, 36H, CH_2), 1.60–1.67 (m, 2H, CH_2), 1.75–1.83 (m, 4H, CH_2), 1.95–2.04 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 2’’’-CH_2), 3.98 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.01 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.59 (t, 2H, *J* = 7.6 Hz, CH_2N), 6.95–6.99 (m, 2H, 2’’-H, 6’’-H), 7.01–7.05 (m, 2H, 3’’’’-H, 5’’’’-H), 7.39–7.51 (m, 2H, 4-H, 5-H), 7.64–7.72 (m, 2H, 2’’’’-H, 6’’’’-H), 8.30–8.37 (m, 2H, 3’’-H, 5’’-H), 8.57 (s, 2H, 4’’’-H, 6’’’-H), 11.11 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.08, 14.13 (CH_3), 22.64, 22.69, 25.9, 26.0, 26.2, 28.9, 29.02, 29.05, 29.13, 29.2, 29.31, 29.35, 29.37, 29.57, 29.60, 29.64, 29.67, 30.2, 30.4, 30.8, 31.8, 31.9 (CH_2), 50.5 (NCH_2), 67.9, 68.7 (OCH_2), 114.4 (C-2’’, C-6’’), 116.1 (C-3’’’’, C-5’’’’), 120.3, 122.0 (C-4, C-5), 123.3 (C-2’’’’, C-6’’’’), 127.2 (C-1’’’’), 129.4 (C-3’’, C-5’’), 130.2 (C-4’’), 132.1 (C-5’’’),136.8 (C-2), 157.0 (C-4’’’, C-6’’’), 160.5 (C-4’’’’), 161.1 (C-1’’), 162.5 (C-2’’’) ppm. MS (ESI): *m/z* = 723.6 $[M–Br]^+$, 555.4, 439.4, 397.3, 355.3, 329.3, 285.2. HRMS (ESI): *m/z*

calcd. for $C_{47}H_{71}N_4O_2^+$: 723.5572, found: 723.5573 $[M–Br]^+$. DSC: Cr_1 54 °C [0.8 kJ/mol] Cr_2 158 °C [37.8 kJ/mol] I (2nd heating), I 130 °C [-36.7 kJ/mol] Cr_2 58 °C [-0.9 kJ/mol] Cr_1 (2nd cooling).

1-{8-[4-[5-Decylpyrimidin-2-yl)phenoxy]octyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide ((*p*-8(10,8)). From *p*-**17**(10,8) (0.10 mmol, 50.0 mg) and **13d** (0.30 mmol, 99.0 mg), 72 h; yield: 74.0 mg (89%). Found: C, 70.75; H, 9.02; N, 6.49%. $C_{49}H_{75}BrN_4O_2$ requires C, 70.73; H, 9.09; N, 6.73; M, 803.99. FT-IR (ATR): ν_{max}/cm^{-1} 3041 (w), 2921 (s), 2853 (m), 1608 (m), 1584 (m), 1568 (w), 1551 (w), 1515 (m), 1467 (m), 1428 (s), 1308 (m), 1256 (s), 1166 (m), 1074 (m), 1018 (m), 846 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 6H *J* = 6.3 Hz, 2 x CH_3), 1.20–1.53 (m, 40H, CH_2), 1.59–1.69 (m, 2H, CH_2), 1.75–1.84 (m, 4H, CH_2), 1.95–2.05 (m, 2H, CH_2), 2.56–2.63 (m, 2H, 2’’’-CH_2), 3.98 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.02 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.59 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.94–6.99 (m, 2H, 2’’-H, 6’’-H), 7.00–7.06 (m, 2H, 3’’’’-H, 5’’’’-H), 7.36–7.51 (m, 2H, 4-H, 5-H), 7.64–7.71 (m, 2H, 2’’’’-H, 6’’’’-H), 8.32–8.39 (m, 2H, 3’’-H, 5’’-H), 8.59 (s, 2H, 4’’’-H, 6’’’-H), 11.13 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.67, 22.69, 25.9, 26.0, 26.1, 28.9, 29.00, 29.04, 29.1, 29.30, 29.35, 29.5, 29.56, 29.60, 29.64, 29.7, 30.1, 30.4, 30.8, 31.88, 31.92 (CH_2), 50.5 (NCH_2), 67.9, 68.7 (OCH_2), 114.5 (C-2’’, C-6’’), 116.1 (C-3’’’’, C-5’’’’), 120.3, 122.0 (C-4, C-5), 123.3 (C-2’’’’, C-6’’’’), 127.2 (C-1’’’’), 129.5 (C-3’’, C-5’’), 130.2 (C-4’’), 132.2 (C-2), 157.0 (C-4’’’, C-6’’’), 160.5 (C-4’’’’), 161.3 (C-1’’), 162.5 (C-2’’’) ppm. MS (ESI): *m/z* = 751.6 $[M–Br]^+$, 583.4, 423.4, 397.3, 355.3, 329.3, 271.2, 161.1, 133.1. HRMS (ESI): *m/z* calcd. for $C_{49}H_{75}N_4O_2^+$: 751.5885, found: 751.5878 $[M–Br]^+$. DSC: Cr 164 °C [40.7 kJ/mol] I (2nd heating), I 142 °C [-1.3 kJ/mol] SmA 137 °C [-38.6 kJ/mol] Cr (2nd cooling).

1-{4-[4-[5-Dodecylpyrimidin-2-yl)phenoxy]octyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide (*p*-8(12,8)). From *p*-**17**(12,8) (94.0 μmol, 50.0 mg) and **13d** (0.94 mmol, 308 mg), 120 h, in DMF; yield: 52.0 mg (64%). Found: C, 71.22; H, 9.26; N, 6.51%. $C_{51}H_{79}BrN_4O_2$ requires C, 71.20; H, 9.22; N, 6.39%; M, 860.1. FT-IR (ATR): ν_{max}/cm^{-1} 3042 (w), 2920 (s), 2852 (m), 1608 (m), 1584 (m), 1568 (w), 1551 (w), 1514 (m), 1467 (m), 1428 (s), 1398 (w), 1327 (w), 1256 (s), 1212 (w), 1183 (w), 1167 (m), 1106 (w), 1074 (w), 1019 (w), 958 (w), 906 (w), 846 (m), 816 (w), 800 (w), 739 (w), 722 (w). δ_H (250 MHz, $CDCl_3$) 0.75–0.84 (m, 6H, CH_3), 1.11–1.45 (m, 44H, CH_2), 1.51–1.62 (m, 2H, 5’’’-$CH_2C\mathit{H}_2$), 1.68–1.77 (m, 2H, $OCH_2C\mathit{H}_2$), 1.87–2.00 (m, 2H, $NC\mathit{H}_2$), 2.47–2.57 (m, 2H, 5’’’-H), 3.90 (t, 2H, *J* = 6.6 Hz, OCH_2), 3.94 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.51 (t, 2H, *J* = 7.6 Hz, NCH_2), 6.86–6.92 (m, 2H, 3’’-H, 5’’-H), 6.93–7.00 (m, 2H, 3’’’’-H, 5’’’’-H), 7.32–7.46 (m, 2H, 4-H, 5-H), 7.57–7.64 (m, 2H, 2’’’’-H, 6’’’’-H), 8.24–8.30 (m, 2H, 2’’-H, 6’’-H), 8.49 (s, 2H, 4’’’-H, 6’’’-H), 11.07

(s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.0, 26.2, 28.9, 29.0, 29.1, 29.4, 29.5, 29.6, 29.7, 30.2, 30.4, 30.8, 31.9 (CH_2), 50.5 (NCH_2), 67.9, 68.7 (2 x OCH_2), 114.4 (C-3'', C-5''), 116.1 (C-3'''', C-5''''), 120.4, 122.1 (C-4, C-5), 123.3 (C-2'''', C-6''''), 127.1 (C-1'''', C-5''', C-1''), 129.4 (C-2'', C-6''), 130.2, 132.1 (C-1'''', C-5''', C-1''), 136.4 (C-2), 157.0 (C-4''', C-6'''), 160.5, 161.1 (C-4'', C-4''''), 162.5 (C-2''') ppm. MS (ESI): *m/z* = 779.6 $[M–Br]^+$. HRMS (ESI): *m/z* calcd. for $C_{51}H_{79}N_4O_2^+$: 779.6198, found: 779.6199 $[M–Br]^+$. DSC: Cr 99 °C [6.9 kJ/mol] I (2nd heating), I 97 °C [-8.2 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-{4-[3-(5-dodecylpyrimidin-2-yl)phenoxy]butyl}imidazolium bromide ((*m*-7(12,4)). From *m*-**17**(12,4) (0.17 mmol, 80.0 mg) and **13c** (1.70 mmol, 397 mg), 96 h; yield: 95.0 mg (79%). Found: C, 68.95; H, 9.34; N, 7.68%. $C_{41}H_{67}BrN_4O$ requires C, 69.17; H, 9.49; N, 7.87%; M, 711.90. FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2853 (s), 1586 (m), 1544 (m), 1446 (m), 1424 (s), 1327 (m), 1290 (m), 1208 (m), 1163 (m), 1039 (w), 904 (s), 782 (m), 726 (s). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 6H, *J* = 6.9 Hz, 2 x CH_3), 1.18–1.41 (m, 36H, CH_2), 1.60–1.70 (m, 2H, CH_2), 1.86–1.97 (m, 4H, CH_2), 2.16–2.26 (m, 2H, CH_2), 2.58–2.65 (m, 2H, 5'''-CH_2), 4.13 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.31 (t, 2H, *J* = 7.6 Hz, NCH_2), 4.53 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.96–7.01 (m, 1H, 6''-H), 7.24–7.34 (m, 2H, 4-H, 5-H), 7.38 (t, 1H, *J* = 7.9, 5''-H), 7.90–7.94 (s, 1H, 2''-H), 7.99–8.04 (m, 1H, 4''-H), 8.61 (s, 2H, 4'''-H, 6'''-H), 10.79 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.3, 26.6, 27.5, 28.9, 29.1, 29.33, 29.34, 29.4, 29.5, 29.53, 29.60, 29.61, 29.64, 30.2, 30.3, 30.8, 31.90, 31.91 (CH_2), 49.9 (CH_2N), 50.2 (NCH_2), 67.1 (OCH_2), 113.2 (C-2''), 117.0 (C-6''), 120.7 (C-4''), 121.4, 121.8 (C-4, C-5), 129.7 (C-5''), 133.3 (C-2), 137.8 (C-3''), 139.2 (C-5'''), 157.0 (C-4''', C-6'''), 158.9 (C-1''), 162.2 (C-2''') ppm. MS (ESI): *m/z* = 631.5 $[M–Br]^+$, 395.3, 353.3, 325.3, 291.3, 237.2. HRMS (ESI): *m/z* calcd. for $C_{41}H_{67}N_4O^+$: 631.5309, found: 631.5316 $[M–Br]^+$. DSC: Cr 55 °C [0.9 kJ/mol] SmA 88 °C [1.5 kJ/mol] I (2nd heating), I 88 °C [-1.6 kJ/mol] SmA (2nd cooling); no recrystallization was observed upon cooling.

3-Dodecyl-1-{8-[3-(5-dodecylpyrimidin-2-yl)phenoxy]octyl}imidazolium bromide (*m*-7(12,8)). From *m*-**17**(12,8) (0.15 mmol, 80.0 mg) and **13c** (1.50 mmol, 355 mg), 96 h; yield: 96.0 mg (83%). Found: C, 69.26; H, 9.61; N, 7.07%. $C_{45}H_{75}BrN_4O$ requires C, 70.37; H, 9.84; N, 7.30%; M, 768.01. FT-IR (ATR): ν_{max}/cm^{-1} 2921 (s), 2851 (s), 1584 (m), 1543 (m), 1447 (m), 1426 (s), 1328 (m), 1297 (m), 1207 (m), 1164 (m), 1044 (w), 902 (s), 779 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 6H, *J* = 6.6 Hz, 2 x CH_3), 1.19–1.42 (m, 38H, CH_2), 1.44–1.53 (m, 2H, CH_2), 1.61–1.69 (m, 2H, CH_2), 1.75–1.84 (m, 2H, CH_2), 1.87–1.98 (m, 4H, CH_2), 2.60–2.65 (m, 2H, 5'''-CH_2), 4.06 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.34 (t, 2H, *J* = 7.3 Hz, NCH_2),

4.37 (t, 2H, *J* = 7.6 Hz, CH_2N), 6.98–7.03 (m, 1H, 6''-H), 7.19–7.24 (m, 2H, 4-H, 5-H), 7.38 (t, 1H, *J* = 7.9, 5''-H), 7.93–7.97 (s, 1H, 2''-H), 7.98–8.02 (m, 1H, 4''-H), 8.62 (s, 2H, 4'''-H, 6'''-H), 10.88 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.1, 26.3, 28.8, 28.9, 29.1, 29.2, 29.3, 29.48, 29.53, 29.59, 29.61, 29.64, 30.2, 30.3, 30.8, 31.91 (CH_2), 50.18 (CH_2N), 50.22 (NCH_2), 67.9 (OCH_2), 113.1 (C-2''), 117.3 (C-6''), 120.3 (C-4''), 121.30, 121.33 (C-4, C-5), 129.6 (C-5''), 133.1 (C-2), 137.8 (C-3''), 139.1 (C-5'''), 157.0 (C-4''', C-6'''), 159.5 (C-1''), 162.4 (C-2''') ppm. MS (ESI): *m/z* = 687.6 $[M–Br]^+$, 519.4, 451.4, 341.3, 305.3, 277.3, 237.2, 207.2, 179.2, 137.1. HRMS (ESI): *m/z* ber. für $C_{45}H_{75}N_4O^+$: 687.5935, gef.: 687.5920$[M–Br]^+$. DSC: Cr_1 46 °C [30.7 kJ/mol] Cr_2 50 °C [1.7 kJ/mol] SmA 54 °C [3.3 kJ/mol] I (2nd heating), I 55 °C [-2.0 kJ/mol] SmA 34 °C [-51.4 kJ/mol] Cr (2nd cooling).

1-{4-[3-[5-Dodecylpyrimidin-2-yl)phenoxy]butyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide ((*m*-8(12,4)). From *m*-**17**(12,4) (0.17 mmol, 80.0 mg) and **13d** (0.51 mmol, 168 mg), 72 h; yield: 131 mg (97%). Found: C, 69.63; H, 8.74; N, 6.81%. $C_{47}H_{71}BrN_4O_3$ requires C, 70.21; H, 8.90; N, 6.97%; M, 803.99. FT-IR (ATR): ν_{max}/cm^{-1} 3097 (w), 2955 (m), 2921 (s), 2851 (s), 1608 (w), 1587 (m), 1547 (m), 1514 (m), 1467 (m), 1451 (m), 1250 (s), 1207 (m), 1191 (m), 1076 (m), 1048 (m), 907 (m), 834 (m), 730 (s). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.19–1.49 (m, 36H, CH_2), 1.60–1.68 (m, 2H, CH_2), 1.75–1.83 (m, 2H, CH_2), 1.92–2.02 (m, 2H, CH_2), 2.22–2.32 (m, 2H, CH_2), 2.58–2.65 (m, 2H, 5'''-CH_2), 3.97 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.14 (t, 2H, *J* = 6.0 Hz, OCH_2), 4.73 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.96–7.00 (m, 1H, 6''-H), 7.00–7.05 (m, 2H, 3''''-H, 5''''-H), 7.37 (t, 1H, *J* = 7.9, 5''-H), 7.46–7.54 (m, 2H, 4-H, 5-H), 7.61–7.67 (m, 2H, 2''''-H, 6''''-H), 7.90–7.95 (s, 1H, 2''-H), 7.98–8.04 (m, 1H, 4''-H), 8.61 (s, 2H, 4'''-H, 6'''-H), 11.07 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.0, 27.5, 29.1, 29.4, 29.54, 29.57, 29.62, 29.64, 29.7, 30.2, 30.8, 31.90 (CH_2), 50.2 (CH_2N), 67.1, 68.7 (2 x OCH_2), 113.3 (C-2''), 116.1 (C-3'''', C-5''''), 117.0 (C-6''), 120.4 (C-4''), 120.6, 122.3 (C-4, C-5), 123.3 (C-2'''', C-6''''), 127.1 (C-1''''), 129.7 (C-5''), 133.2 (C-2), 139.2 (C-5'''), 157.0 (C-4''', C-6'''), 158.9 (C-1''), 160.5 (C-4''''), 162.2 (C-2''') ppm. MS (ESI): *m/z* = 723.6 $[M–Br]^+$, 395.3, 353.3, 325.3.7. HRMS (ESI): *m/z* calcd. for $C_{47}H_{71}N_4O_2^+$: 723.5572, found: 723.5569 $[M–Br]^+$. DSC: Cr_1 42 °C [1.8 kJ/mol] Cr_2 58 °C [5.0 kJ/mol] Cr_3 64 °C [4.4 kJ/mol] Cr_4 79 °C [1.8 kJ/mol] Cr_5 104 °C [8.3 kJ/mol] Cr_6 125 °C [11.7 kJ/mol] SmA 157 °C [1.2 kJ/mol] I (2nd heating), I 156 °C [-2.5 kJ/mol] SmA 129 °C [-13.7 kJ/mol] Cr_4 63 °C [-13.8 kJ/mol] Cr_3 56 °C [-5.1 kJ/mol] Cr_2 29 °C [-0.9 kJ/mol] Cr_1 (2nd cooling).

1-{8-[3-[5-Dodecylpyrimidin-2-yl)phenoxy]octyl}-3-[4-(dodecyloxy)phenyl]imidazolium bromide (*m*-8(12,8)). From *m*-**17**(12,8) (0.15 mmol, 80.0 mg) and **13d** (0.45 mmol, 148 mg), 72 h; yield: 116 mg (90%). Found: C, 70.78; H, 9.13; N, 6.33%. $C_{51}H_{79}BrN_4O_2$ requires C, 71.22; H, 9.26; N, 6.51%; M, 860.10. FT-IR (ATR): ν_{max}/cm^{-1} 3053 (w), 2924 (s), 2853 (s), 1586 (m), 1544 (m), 1514 (m), 1467 (m), 1447 (m), 1327 (w), 1255 (s), 1208 (m), 1074 (m), 1029 (m), 833 (m), 782 (m), 653 (m) cm^{-1}. δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 6.9 Hz, CH_3), 0.89 (t, 3H, *J* = 6.9 Hz, CH_3), 1.19–1.52 (m, 44H, CH_2), 1.60–1.69 (m, 2H, CH_2), 1.75–1.83 (m, 4H, CH_2), 1.94–2.04 (m, 2H, CH_2), 2.58–2.65 (m, 2H, 5’’’-CH_2), 3.97 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.05 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.58 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.98–7.01 (m, 1H, 6’’-H), 7.00–7.05 (m, 2H, 3’’’’-H, 5’’’’-H), 7.37 (t, 1H, *J* = 8.5, 5’’-H), 7.41–7.54 (m, 2H, 4-H, 5-H), 7.65–7.71 (m, 2H, 2’’’’-H, 6’’’’-H), 7.92–7.96 (s, 1H, 2’’-H), 7.97–8.01 (m, 1H, 4’’-H), 8.62 (s, 2H, 4’’’-H, 6’’’-H), 11.07 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.9, 26.1, 28.9, 29.1, 29.2, 29.4, 29.5, 29.57, 29.61, 29.64, 30.2, 30.3, 30.8, 31.9 (CH_2), 50.5 (CH_2N), 67.9, 68.7 (2 x OCH_2), 113.3 (C-2’’), 116.1 (C-3’’’’, C-5’’’’), 117.3 (C-6’’), 120.2 (C-4’’), 120.4, 122.1 (C-4, C-5), 123.3 (C-2’’’’, C-6’’’’), 127.2 (C-1’’’’), 129.6 (C-5’’), 133.1 (C-2), 139.1 (C-5’’’), 157.0 (C-4’’’, C-6’’’), 159.5 (C-1’’), 160.4 (C-4’’’’), 162.4 (C-2’’’) ppm. MS (ESI): *m/z* = 779.6 $[M–Br]^+$, 451.4, 397.3, 329.3, 300.1, 161.1. HRMS (ESI): *m/z* $[M–Br]^+$ calcd. for $C_{51}H_{79}N_4O_2^+$: 779.6198, found: 779.6198. DSC: Cr_1 10 °C [1.0 kJ/mol] Cr_2 41 °C [-9.4 kJ/mol] Cr_3 91 °C [21.0 kJ/mol] SmA 108 °C [1.5 kJ/mol] I (2nd heating); I 109 °C [-1.8 kJ/mol] SmA 28 °C [-1.2 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-(4-{[2-tetradecylpyrimidin-5-yl)carbonyl]oxy}butyl)imidazolium bromide (22(4)). From **20**(4) (0.13 mmol, 60.0 mg) and **13c** (1.30 mmol, 307 mg), 96 h; yield: 59.0 mg (66%). Found: C, 65.95; H, 9.66; N, 7.95%. $C_{38}H_{67}BrN_4O_2$ requires C, 65.97; H, 9.76; N, 8.10%; M, 691.87. FT-IR (ATR): ν_{max}/cm^{-1} 2956 (m), 2922 (s), 2853 (m), 1722 (s), 1590 (s), 1562 (m), 1466 (m), 1442 (m), 1389 (m), 1291 (s), 1163 (m), 1137 (m), 1038 (w), 722 (m), 647 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, 2 x CH_3), 1.18–1.42 (m, 40H, CH_2), 1.79–1.87 (m, 2H, CH_2), 1.88–1.97 (m, 4H, CH_2), 2.08–2.16 (m, 2H, CH_2), 2.99–3.06 (m, 2H, 2-CH_2), 4.31 (t, 2H, *J* = 7.6 Hz, CH_2N), 4.44 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.55 (t, 2H, *J* = 7.6 Hz, CH_2N), 7.22–7.36 (m, 2H, 4-H, 5-H), 9.17 (s, 2H, 4’’-H, 6’’-H), 10.85 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.6, 26.3, 27.1, 28.6, 29.0, 29.3, 29.36, 29.39, 29.42, 29.48, 29.53, 29.59, 29.64, 29.66, 29.69, 30.2, 31.90, 31.93 (CH_2), 39.8 (2’’-CH_2), 49.5, 50.4 (CH_2N, NCH_2), 64.3 (OCH_2), 121.1 (C-5’’), 121.4, 121.6 (C-4, C-5), 138.1 (C-2), 158.1 (C-4’’, C-6’’), 164.0 (CO_2), 175.5 (C-2’’) ppm. MS (ESI): *m/z* = 611.5 $[M–Br]^+$, 443.3, 375.3, 321.3, 291.3, 237.2, 123.1. HRMS (ESI): *m/z* calcd. for $C_{38}H_{67}N_4O_2^+$: 611.5259, found:

611.5265 $[M–Br]^+$. DSC: Cr 41 °C [18.7 kJ/mol] SmA 164 °C [1.8 kJ/mol] I (2nd heating), I 164 °C [-2.1 kJ/mol] SmA 33 °C [-23.3 kJ/mol] Cr (2nd cooling).

3-Dodecyl-1-(8-{[2-tetradecylpyrimidin-5-yl)carbonyl]oxy}octyl)imidazolium bromide (22(8)). From **20**(8) (0.137 mmol, 70.0 mg) and **13c** (0.27 mmol, 64.0 mg), 96 h; yield: 64.0 mg (62%). Found: C, 67.82; H, 9.77; N, 7.08%. $C_{42}H_{75}BrN_4O_2$ requires C, 67.44; H, 10.11; N, 7.49%; M, 747.97. FT-IR (ATR): ν_{max}/cm^{-1} 3051 (w), 2920 (s), 2851 (m), 1711 (s), 1590 (s), 1553 (m), 1466 (m), 1445 (m), 1389 (m), 1289 (s), 1165 (m), 1136 (m), 1037 (w), 729 (m), 648 (m). δ_H (500 MHz, $CDCl_3$) 0.90 (t, 3H, *J* = 6.9 Hz, 2 x CH_3), 1.22–1.48 (m, 40H, CH_2), 1.75–1.89 (m, 4H, CH_2), 1.91–2.01 (m, 4H, CH_2), 3.02–3.07 (m, 2H, 2-CH_2), 4.37 (2 x t, 4H, *J* = 7.1 Hz, CH_2N, NCH_2), 4.40 (t, 2H, *J* = 7.6 Hz, OCH_2), 7.24–7.28 (m, 2H, 4-H, 5-H), 9.20 (s, 2H, 4''-H, 6''-H), 10.94 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.8, 26.2, 26.3, 28.5, 28.6, 28.86, 28.95, 29.0, 29.33, 29.38, 29.42, 29.49, 29.53, 29.59, 29.63, 29.66, 29.69, 30.3, 31.91, 31.93 (CH_2), 39.8 (2''-CH_2), 50.2, 50.3 (CH_2N, NCH_2), 65.6 (OCH_2), 121.3 (C-5''), 121.34, 121.5 (C-4, C-5), 138.1 (C-2), 158.1 (C-4'', C-6''), 164.1 (CO_2), 175.3 (C-2'') ppm. MS (ESI): *m/z* = 667.6 $[M–Br]^+$, 499.4, 347.3, 321.3. HRMS (ESI): *m/z* calcd. for $C_{42}H_{75}N_4O_2^+$: 667.5885, found: 667.5872 $[M–Br]^+$. DSC: Cr 55 °C [35.2 kJ/mol] SmA 76 °C [1.3 kJ/mol] I (2nd heating), I 77 °C [-1.4 kJ/mol] SmA 38 °C [-45.0 kJ/mol] Cr (2nd cooling).

1-(4-{[(2-Tetradecylpyrimidin-5-yl)carbonyl]oxy}butyl)-3-[4-(dodecyloxy)phenyl]-imidazolium bromide (23(4)). From **20**(4) (0.21 mmol, 95.0 mg) and **13d** (0.63 mmol, 206 mg), 72 h; yield: 148 mg (90%). FT-IR (ATR): ν_{max}/cm^{-1} 2956 (m), 2921 (s), 2852 (m), 1720 (s), 1591 (s), 1553 (m), 1514 (s), 1467 (m), 1443 (m), 1390 (m), 1293 (s), 1256 (s), 1137 (m), 1039 (w), 905 (s), 729 (s), 649 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 7.0 Hz, 2 x CH_3), 1.18–1.51 (m, 40H, CH_2), 1.71–1.86 (m, 4H, CH_2), 1.92–2.04 (m, 2H, CH_2), 2.12–2.23 (m, 2H, CH_2), 2.94–3.06 (m, 2H, 2-CH_2), 3.98 (t, 2H, *J* = 7.9 Hz, CH_2N), 4.46 (t, 2H, *J* = 6.3 Hz, OCH_2), 4.75 (t, 2H, *J* = 7.3 Hz, CH_2N), 6.99–7.09 (m, 2H, 3'''-H, 5'''-H), 7.50–7.59 (m, 2H, 4-H, 5-H), 7.62–7.69 (m, 2H, 2'''-H, 6'''-H), 9.17 (s, 2H, 4''-H, 6''-H), 11.10 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.6, 27.0, 27.2, 28.6, 29.1, 29.36, 29.38, 29.42, 29.5, 29.57, 29.60, 29.66, 29.68, 31.9 (CH_2), 39.8 (2''-CH_2), 49.8 (CH_2N), 64.4, 68.7 (OCH_2), 116.1 (C-3''', C-5'''), 120.5 (C-5''), 121.1, 122.3 (C-4, C-5), 123.3 (C-2''', C-6'''), 127.1 (C-1'''), 136.4 (C-2), 158.1 (C-4'', C-6''), 160.6 (C-4'''), 164.0 (CO_2), 175.5 (C-2'') ppm. MS (ESI): *m/z* = 703.6 $[M–Br]^+$, 383.3, 329.3, 303.2, 215.1, 161.1, 123.1. HRMS (ESI): *m/z* calcd. for $C_{44}H_{71}N_4O_3^+$: 703.5521, found: 703.5520 $[M–Br]^+$. DSC: Cr 77 °C [78.2 kJ/mol] SmA 229 °C [2.6 kJ/mol] I (1st heating); decomposition upon clearing.

1-(8-{[(2-Tetradecylpyrimidin-5-yl)carbonyl]oxy}octyl)-3-[4-(dodecyloxy)phenyl]imidazolium bromide (23(8)). From **20**(8) (0.14 mmol, 70.0 mg) and **13d** (0.27 mmol, 90.0 mg), 72 h; yield: 105 mg (91%). Found: C, 68.61; H, 9.25; N, 6.61%. $C_{48}H_{79}BrN_4O_3$ requires C, 68.63; H, 9.48; N, 6.67%; M, 840.07. FT-IR (ATR): ν_{max}/cm^{-1} 3046 (w), 2920 (s), 2851 (m), 1720 (s), 1590 (s), 1551 (m), 1516 (w), 1469 (m), 1442 (m), 1394 (w), 1289 (s), 1257 (m), 1136 (m), 1075 (w). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 6H, *J* = 6.9 Hz, 2 x CH_3), 1.19–1.50 (m, 48H, CH_2), 1.73–1.87 (m, 6H, CH_2), 1.96–2.05 (m, 2H, CH_2), 2.98–3.06 (m, 2H, 2-CH_2), 3.98 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.35 (t, 2H, *J* = 6.9 Hz, NCH_2), 4.60 (t, 2H, *J* = 7.3 Hz, CH_2N), 7.01–7.06 (m, 2H, 3’’’-H, 5’’’-H), 7.39–7.52 (m, 2H, 4-H, 5-H), 7.66–7.70 (m, 2H, 2’’’-H, 6’’’-H), 9.17 (s, 2H, 4’’-H, 6’’-H), 11.15 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.8, 26.0, 26.2, 28.5, 28.6, 28.91, 28.94, 29.1, 29.36, 29.42, 29.5, 29.57, 29.60, 29.62, 29.66, 29.68, 30.4, 31.90 (CH_2), 39.8 (2’’-CH_2), 50.4 (CH_2N), 65.6, 68.7 (2 x OCH_2), 116.1 (C-3’’’, C-5’’’), 120.3 (C-5’’), 121.5, 122.0 (C-4, C-5), 123.3 (C-2’’’, C-6’’’), 127.1 (C-1’’’), 136.5 (C-2), 158.1 (C-4’’, C-6’’), 160.5 (C-4’’’), 164.1 (CO_2), 175.3 (C-2’’) ppm. MS (ESI): *m/z* = 759.6 $[M–Br]^+$, 591.4, 439.4, 329.3. HRMS (ESI): *m/z* calcd. for $C_{48}H_{79}N_4O_3^+$: 759.6147, found: 759.6152 $[M–Br]^+$. DSC: Cr_1 31 °C [7.9 kJ/mol] Cr_2 101 °C [18.2 kJ/mol] SmA 172 °C [2.6 kJ/mol] I (2nd heating); I 172 °C [-2.3 kJ/mol] SmA 76 °C [-14.4 kJ/mol] 36 °C [-8.3 kJ/mol] (2nd cooling).

General procedure for the preparation of trifluoromethanesulfonates 24(m)–26(m). To a solution of the respective compound **21(m)**–**23(m)** (1.00 mmol) in acetonitrile (5 mL) at room temperature was added potassium trifluoromethanesulfonate (1.10 mmol), whereby colourless KBr precipitated. The reaction mixture was stirred for 2 h (tlc control). After completion of the reaction, the solvent was removed under vacuum. The residue was taken up in CH_2Cl_2 (5 mL) and filtered. The products were obtained as colourless solids without further purification.

3-Methyl-1-(4-{[(2-tetradecylpyrimidin-5-yl)carbonyl]oxy}butyl)imidazolium trifluoromethanesulfonate (24(4)). From **21**(4) (0.019 mmol, 10.0 mg); yield: 12.0 mg (quant.). FT-IR (ATR): ν_{max}/cm^{-1} 2922 (s), 2853 (m), 1723 (s), 1590 (s), 1443 (m), 1390 (m), 1257 (s), 1163 (m), 1031 (w), 904 (s), 727 (s), 649 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.9 Hz, CH_3), 1.19–1.41 (m, 26H, CH_2), 1.77–1.92 (m, 4H, CH_2), 2.03–2.11 (m, 2H, CH_2), 2.98–3.05 (m, 2H, 2-CH_2), 4.00 (s, 3H, NCH_3), 4.34 (t, 2H, *J* = 7.3 Hz, CH_2N), 4.41 (t, 2H, *J* = 6.3 Hz, OCH_2), 7.29–7.38 (m, 2H, 4-H, 5-H), 9.15 (s, 2H, 4’’-H, 6’’-H), 9.30 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.4, 26.8, 28.6, 29.2, 29.36, 29.39, 29.42, 29.5, 29.6, 29.66, 29.69, 31.9 (CH_2), 36.6 (NCH_3), 39.8 (2’’-CH_2), 49.6 (CH_2N), 64.2 (OCH_2),

121.1, 121.9 (C-4, C-5), 122.0 (CF_3), 123.4 (C-5''), 137.5 (C-2), 158.1 (C-4'', C-6''), 164.0 (CO_2), 175.5 (C-2'') ppm. MS (ESI, positive): *m/z* = 457.4 $[M– CF_3SO_3]^+$, 375.3, 321.3, 303.2, 177.1, 137.1. MS (ESI, negative): *m/z* = 149.0 $[CF_3SO_3]^–$, 99.0, 80.0. HRMS (ESI, positive): *m/z* calcd. for $C_{27}H_{45}N_4O_2^+$: 457.3537, found: 457.3538 $[M– CF_3SO_3]^+$. HRMS (ESI, negative): *m/z* calcd. for $CF_3O_3S^–$: 148.9515, found: 148.9524 $[CF_3SO_3]^–$. DSC: Cr_1 0 °C [1.7 kJ/mol] Cr_2 35 °C [19.2 kJ/mol] SmA 142 °C [0.6 kJ/mol] I (2nd heating), I 145 °C [-0.6 kJ/mol] SmA 36 °C [-18.3 kJ/mol] Cr_2 3 °C [-1.2 kJ/mol] Cr_1 (2nd cooling).

3-Dodecyl-1-(4-{[(2-tetradecylpyrimidin-5-yl)carbonyl]oxy}butyl)imidazolium trifluoromethanesulfonate (25(4)). From **22**(4) (0.043 mmol, 30.0 mg); yield: 33.0 mg (quant.). FT-IR (ATR): ν_{max}/cm^{-1} 2923 (s), 2854 (m), 1725 (s), 1591 (s), 1466 (m), 1389 (m), 1259 (s), 1161 (m), 1031 (m), 723 (m), 649 (m). δ_H (500 MHz, $CDCl_3$) 0.88 (t, 3H, *J* = 6.6 Hz, 2 x CH_3), 1.18–1.42 (m, 40H, CH_2), 1.78–1.94 (m, 6H, CH_2), 2.02–2.11 (m, 2H, CH_2), 2.97–3.06 (m, 2H, 2-CH_2), 4.22 (t, 2H, *J* = 7.6 Hz, CH_2N), 4.38 (t, 2H, *J* = 7.6 Hz, CH_2N), 4.42 (t, 2H, *J* = 6.6 Hz, OCH_2), 7.22–7.34 (m, 2H, 4-H, 5-H), 9.16 (s, 2H, 4''-H, 6''-H), 9.47 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.5, 26.2, 26.9, 28.6, 28.9, 29.3, 29.36, 29.39, 29.42, 29.46, 29.53, 29.57, 29.64, 29.66, 29.68, 30.1, 31.89, 31.92 (CH_2), 39.8 (2''-CH_2), 49.6, 50.4 (CH_2N, NCH_2), 64.2 (OCH_2), 121.1 (C-5''), 121.7 (CF_3), 121.80, 121.83 (C-4, C-5), 137.2 (C-2), 158.1 (C-4'', C-6''), 164.0 (CO_2), 175.5 (C-2'') ppm. MS (ESI, positive): *m/z* = 611.5 $[M– CF_3SO_3]^+$, 443.3, 375.3, 321.3, 291.3, 237.2, 123.1. MS (ESI, negative): *m/z* = 149.0 $[CF_3SO_3]^–$, 99.0, 80.0. HRMS (ESI, positive): *m/z* calcd. for $C_{38}H_{67}N_4O_2^+$: 611.5259, found: 611.5247 $[M– CF_3SO_3]^+$. HRMS (ESI, negative): *m/z* calcd. for $CF_3O_3S^–$: 148.9515, found: 148.9525 $[CF_3SO_3]^–$. DSC: Cr_1 6 °C [8.2 kJ/mol] Cr_2 18 °C [1.7 kJ/mol] Cr_3 25 °C [21.0 kJ/mol] SmA 73 °C [1.1 kJ/mol] I (2nd heating); I 73 °C [-1.2 kJ/mol] SmA 23 °C [-19.5 kJ/mol] Cr_3 18 °C [-1.4 kJ/mol] Cr_2 5 °C [-4.0 kJ/mol] Cr_3 (2nd cooling).

3-Dodecyl-1-(8-{[(2-tetradecylpyrimidin-5-yl)carbonyl]oxy}octyl)imidazolium trifluoromethanesulfonate (25(8)). From **22**(8) (0.027 mmol, 20.0 mg); yield: 22.0 mg (quant.), mp 22 °C. FT-IR (ATR): ν_{max}/cm^{-1} 2925 (w), 2854 (m), 1727 (s), 1591 (s), 1466 (m), 1442 (m), 1389 (m), 1258 (s), 1159 (m), 1031 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 7.0 Hz, 2 x CH_3), 1.19–1.49 (m, 48H, CH_2), 1.72–1.95 (m, 8H, CH_2), 2.99–3.05 (m, 2H, 2-CH_2), 4.23 (t, 2H, *J* = 7.5 Hz, CH_2N), 4.25 (t, 2H, *J* = 7.5 Hz, CH_2N), 4.35 (t, 2H, *J* = 6.7 Hz, OCH_2), 7.23–7.27 (m, 2H, 4-H, 5-H), 9.17 (s, 2H, 4''-H, 6''-H), 9.42 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.69, 22.74, 25.7, 26.1, 26.2, 28.49, 28.58, 28.76, 28.88, 28.91, 29.33, 29.36, 29.38, 29.42, 29.47, 29.52, 29.58, 29.62, 29.65, 29.68, 29.69, 30.1, 31.89,

31.92 (CH_2), 39.8 (2''-CH_2), 50.2, 50.3 (CH_2N, NCH_2), 65.6 (OCH_2), 121.5 (C-5''), 121.7 (C-4, C-5), 137.1 (C-2), 158.1 (C-4'', C-6''), 164.1 (CO_2), 175.3 (C-2'') ppm. MS (ESI, positive): *m/z* = 667.6 [M– CF_3SO_3]$^+$, 499.4, 347.3, 321.3, 237.2, 179.2. MS (ESI, negative): *m/z* = 149.0 [CF_3SO_3]$^-$, 99.0, 80.0. HRMS (ESI, positive): *m/z* calcd. for $C_{42}H_{75}N_4O_2^+$: 667.5885, found: 667.5889 [M– CF_3SO_3]$^+$. HRMS (ESI, negative): *m/z* calcd. for $CF_3O_3S^-$: 148.9515, found: 148.9533 [CF_3SO_3]$^-$.

1-(4-{[(2-Tetradecylpyrimidin-5-yl)carbonyl]oxy}butyl)-3-[4-(dodecyloxy)phenyl]-imidazolium trifluoromethanesulfonate (26(4)). From **23**(4) (0.057 mmol, 45.0 mg); yield: 49.0 mg (quant.). FT-IR (ATR): ν_{max}/cm^{-1} 2920 (s), 2851 (m), 1722 (s), 1591 (s), 1555 (m), 1514 (s), 1468 (m), 1389 (m), 1255 (s), 1159 (m), 1031 (w), 902 (s), 723 (s), 650 (m). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 7.0 Hz, 2 x CH_3), 1.19–1.51 (m, 40H, CH_2), 1.75–1.86 (m, 4H, CH_2), 1.89–1.97 (m, 2H, CH_2), 2.08–2.17 (m, 2H, CH_2), 2.98–3.04 (m, 2H, 2-CH_2), 3.99 (t, 2H, *J* = 6.6 Hz, CH_2N), 4.43 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.52 (t, 2H, *J* = 7.6 Hz, CH_2N), 7.01–7.05 (m, 2H, 3'''-H, 5'''-H), 7.49–7.51 (m, 2H, 2'''-H, 6'''-H), 7.51–7.55 (m, 2H, 4-H, 5-H), 9.16 (s, 2H, 4''-H, 6''-H), 9.67 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.5, 26.0, 27.1, 28.6, 29.1, 29.36, 29.39, 29.42, 29.5, 29.57, 29.60, 29.64, 29.66, 29.68, 31.9 (CH_2), 39.8 (2''-CH_2), 49.9 (CH_2N), 64.3, 68.7 (2 x OCH_2), 116.1 (C-3''', C-5'''), 119.5 (CF_3), 121.1 (C-5''), 121.3, 122.5 (C-4, C-5), 123.5 (C-2''', C-6'''), 127.0 (C-1'''), 135.5 (C-2), 158.1 (C-4'', C-6''), 160.7 (C-4'''), 164.1 (CO_2), 175.5 (C-2'') ppm. MS (ESI, positive): *m/z* = 703.6 [M– CF_3SO_3]$^+$, 535.4, 383.3, 329.3, 303.2, 215.1, 161.1. MS (ESI, negative): *m/z* = 149.0 [CF_3SO_3]$^-$, 99.0, 80.0. HRMS (ESI, positive): *m/z* calcd. for $C_{44}H_{71}N_4O_3^+$: 703.5521, found: 703.5520 [M– CF_3SO_3]$^+$. HRMS (ESI, negative): *m/z* calcd. for $CF_3O_3S^-$: 148.9515, found: 148.9516 [CF_3SO_3]$^-$. DSC: Cr_1 33 °C [24.2 kJ/mol] Cr_2 51 °C [-61.6 kJ/mol] Cr_3 83 °C [81.6 kJ/mol] Cr_4 90 °C [19.0 kJ/mol] SmA 142 °C [2.1 kJ/mol] I (2nd heating); I 143 °C [-2.1 kJ/mol] SmA 26 °C [-13.1 kJ/mol] Cr_2 20 °C [-6.5 kJ/mol] Cr_1 (2nd cooling).

1-(8-{[(2-Tetradecylpyrimidin-5-yl)carbonyl]oxy}octyl)-3-[4-(dodecyloxy)phenyl]imidazolium trifluoromethanesulfonate (26(8)). From **23**(8) (0.071 mmol, 60.0 mg); yield: 65.0 mg (quant.). FT-IR (ATR): ν_{max}/cm^{-1} 2921 (s), 2852 (m), 1725 (s), 1591 (s), 1554 (m), 1514 (s), 1468 (m), 1391 (m), 1256 (s), 1160 (m), 1031 (w). δ_H (500 MHz, $CDCl_3$) 0.87 (t, 3H, *J* = 7.0 Hz, 2 x CH_3), 1.18–1.51 (m, 48H, CH_2), 1.73–1.87 (m, 6H, CH_2), 1.89–1.99 (m, 2H, CH_2), 2.98–3.06 (m, 2H, 2-CH_2), 3.99 (t, 2H, *J* = 6.3 Hz, CH_2N), 4.35 (t, 2H, *J* = 6.6 Hz, OCH_2), 4.39 (t, 2H, *J* = 7.6 Hz, CH_2N), 7.01–7.06 (m, 2H, 3'''-H, 5'''-H), 7.40–7.51 (m, 2H, 2'''-H, 6'''-H), 7.52–7.56 (m, 2H, 4-H, 5-H), 9.17 (s, 2H,

4''-H, 6''-H), 9.61 (s, 1H, 2-H) ppm. δ_C (125 MHz, $CDCl_3$) 14.1 (CH_3), 22.7, 25.7, 26.0, 26.1, 28.5, 28.6, 28.8, 28.9, 29.1, 29.36, 29.42, 29.52, 29.57, 29.60, 29.65, 29.7, 30.2, 31.9 (CH_2), 39.8 (2''-CH_2), 50.6 (CH_2N), 65.6, 68.7 (OCH_2), 116.1 (C-3''', C-5'''), 121.1 (C-5''), 121.4 (CF_3), 121.3, 122.5 (C-4, C-5), 123.5 (C-2''', C-6'''), 127.0 (C-1'''), 135.4 (C-2), 158.1 (C-4'', C-6''), 160.7 (C-4'''), 164.1 (CO_2), 175.3 (C-2'') ppm. MS (ESI, positive): *m/z* = 759.6 $[M- CF_3SO_3]^+$, 591.4, 439.4, 329.3, 271.2, 161.1. MS (ESI, negative): *m/z* = 149.0 $[CF_3SO_3]^-$, 99.0, 80.0. HRMS (ESI, positive): *m/z* calcd. for $C_{48}H_{79}N_4O_3^+$: 759.6147, found: 759.6131 $[M- CF_3SO_3]^+$. HRMS (ESI, negative): *m/z* calcd. for $CF_3O_3S^-$: 148.9515, found: 148.9523 $[CF_3SO_3]^-$. DSC: Cr_1 21 °C [-17.7 kJ/mol] Cr_2 44 °C [36.1 kJ/mol] Cr_3 49 °C [8.5 kJ/mol] SmA 72 °C [1.6 kJ/mol] I (2nd heating); I 72 °C [-1.7 kJ/mol] SmA 29 °C [-16.7 kJ/mol] Cr_2 17 °C [-10.6 kJ/mol] Cr_1 (2nd cooling).

2. X-ray data

Table S1. Temperature dependent layer spacings of compounds **5c**–**f**.

5c		**5d**		**5e**		**5f**	
T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]
130	5355	105	4833	79	5604	83	5990
139	5222	115	4675	83	5702	93	5930
149	5095	124	4574	88	5706	102	5966
158	4987	134	4495	93	5709	112	5971
168	4887	143	4418	98	5727	121	6011
177	4798	153	4354	102	5752	131	6221
187	4708			107	5790	140	6040
196	4638			112	5863	150	5768
206	4568			117	6018	159	5505
215	4514			121	5969	169	5257
225	4469			126	5873	178	5072
				131	5765		
				136	5629		
				140	5479		
				145	5323		
				150	5175		
				154	4960		

Table S2. Temperature dependent layer spacings of compounds **6f**, **7c**–**9c** and **11c**.

6f		**7c**		**8c**		**9c**		**11c**	
T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]
69	6393	107	3602	137	3952	102	4343	29	3500
72	6292	111	3581	142	3923	112	4305	34	3468
75	6131	115	3560	147	3897	121	4260	39	3439
78	5957	118	3541	152	3874	131	4221	43	3410
80	5788	122	3524	156	3818	140	4188	48	3383
83	5623			161	3818	150	4152	53	3357
86	5466			166	3790	159	4115	58	3332
89	5333			171	3763	169	4089	62	3308
92	5216							67	3286
95	5123							72	3265
								77	3243

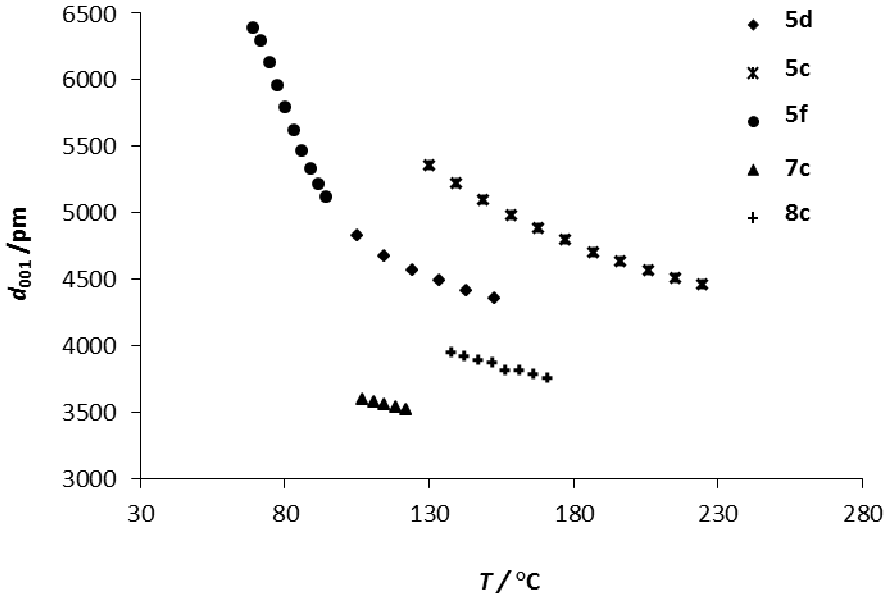


Fig. S1. Temperature dependence of the layer spacing for compounds **5c,d,f**, **7c** and **8c**.

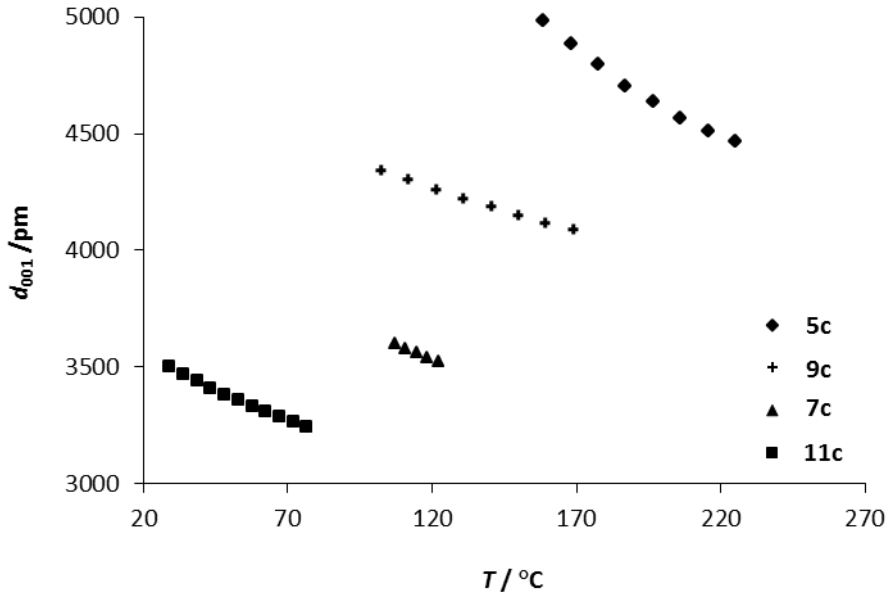


Fig. S2. Influence of different substituted imidazolium head groups (Me vs. $C_{12}H_{25}$) on temperature dependent layer spacings.

Table S3. Temperature dependent layer spacings of compounds **31b** and **32a,b.**

31b		**32a**		**33a**	
T / [°C]	d_{001} / [pm]	T / [°C]	d_{001} / [pm]	T / [°C]	d_{001}
45	4792	28	3291	78	3570
49	4727	32	3268	87	3528
53	4675	36	3249	95	3489
58	4625	40	3232	104	3452
62	4582	45	3216	113	3415
66	4538	49	3202	121	3380
701	4503	53	3189	130	3345
		58	3177		
		62	3165		

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