Supplementary Information:

Self-Assembly of Cyclobis(paraquat-*p*-phenylene)s

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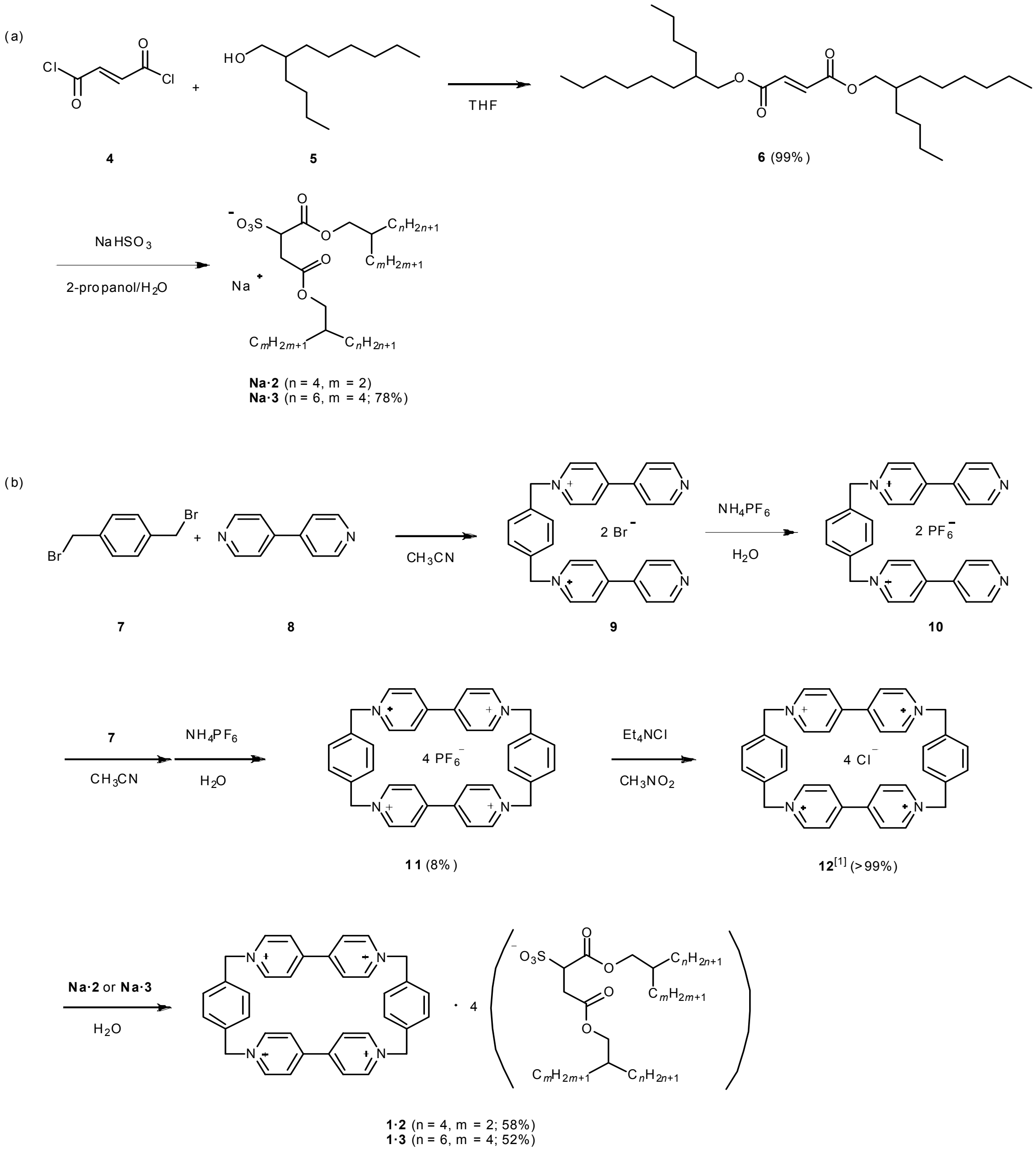
Experimental Details:

General. ^{1}H and ^{13}C{^{1}H} NMR spectra were recorded on a JEOL JNM-LA400 spectrometer. Mass spectra were obtained with a PerSeptive Biosystems Voyager-DE STR spectrometer. Elemental analyses were carried out with a Yanaco MT-6 CHN autocorder. Differential scanning calorimetry (DSC) measurements were performed on a NETZSCH DSC204 Phoenix calorimeter at a scanning rate of 10 °C min^{-1}. A polarizing optical microscope Olympus BH-51 equipped with Mettler FP82 HT hot stage was used for visual observation. X-ray diffraction (XRD) patterns were obtained using a Rigaku RINT-2500 diffractmeter with a heating stage using Ni-filtered CuKα radiation. UV-vis absorption spectra were measured with a JASCO V-670 spectrometer.

Materials and Syntheses. All reagents and solvents were purchased from Aldrich, Tokyo Kasei, Kanto Chemical, or Wako, and used as received. The synthetic routes used to obtain viologen-based cyclophanes **1·2** and **1·3** are shown in Scheme S1.

Viologen-based cyclophane with Cl anions (**12**) were prepared according to the literature.[1] All reactions were performed under an Ar atmosphere in dry solvents, unless otherwise noted.

Scheme S1. Syntheses of (a) surfactant **Na·3** and (b) viologen-based cyclophanes **1·2** and **1·3**.



Di(2-butyloctyl) Fumarate (6). To a stirred solution of **5** (9.32 g, 40 mmol) in THF (80 mL) was dropwisely added **4** (3.06 g, 20 mmol) at 0 °C. After stirred for 24 h at room temperature, the reaction mixture was poured into saturated aqueous $NaHCO_3$ (100 mL), and extracted with ethyl acetate/hexane, three times. The combined organic layers were washed with saturated aqueous $NaHCO_3$ and brine, successively, and dried over anhydrous Na_2SO_4. After filtration and evaporation, the product was purified by column chromatography (silica, $CHCl_3$ as an eluent), and dried under vacuum to provide **6** as a colorless oil (yield = 9.02 g, 99%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 6.85 (s, 2H), 4.12 (d, *J* = 6.0 Hz, 4H), 1.70–1.66 (m, 2H), 1.33–1.25 (m, 32H), 0.92–0.86 (m, 12H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ165.10, 133.51, 68.03, 37.20, 31.72, 31.15, 30.83, 29.51, 28.84, 26.59, 22.88, 22.57, 13.99, 13.94. MS (MALDI): *m/z* 491.02 (calcd $[M{+}K]^+$ = 491.81).

Sodium Di(2-butyloctyl) Sulfosuccinate (Na·3). To a stirred solution of **6** (4.53 g, 10 mmol) in 2-propanol (80 mL; Ar bubbled before use) was dropwisely added aqueous $NaHSO_3$ (0.3 M, 40 mL; Ar bubbled before use) at room temperature. The mixture was stirred for 24 h at 80 °C. After evaporation of the solvent, the residue was dissolved in chloroform and dried over anhydrous Na_2SO_4. After filtration and evaporation, the product was dried under vacuum to afford **Na·3** as a white waxy solid (yield = 4.34 g, 78%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 4.30 (m, 1H), 4.06 (m, 2H), 3.94 (m, 2H), 1.62 (m, 2H), 1.35–1.20 (m, 32H), 0.84 (m, 12 H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 171.53, 169.70, 69.21, 68.04, 61.24, 37.15, 36.96, 33.00, 31.88, 31.01, 30.80, 30.65, 30.38, 29.68, 28.81, 28.76, 28.71, 28.66, 26.66, 26.62, 26.56, 22.99, 22.96, 22.71, 22.67, 14.08, 14.06. MS (MALDI): *m/z* 579.24 (calcd $[M{+}Na]^+$ = 579.76).

Cyclobis(paraquat-*p*-phenylene) Tetrakis[Di(2-ethylhexyl) Sulfosuccinate] (1·2). To a stirred solution of **12** (132 mg, 0.2 mmol) in water (10 mL) was added a solution of aerosol OT **Na·2** (445 mg, 1.0 mmol) in water/methanol (5:1, 5 mL) at room temperature. The formed precipitate was filtered off, washed with water, and recrystallized from methanol/water to give **1·2** as a white solid (254 mg, 58%). ^{1}H NMR (400 MHz, DMSO-*d*$_6$): δ 9.44 (d, *J* = 6.4 Hz, 8H), 8.65 (d, *J* = 6.4 Hz, 8H), 7.12 (s, 8H), 5.81 (s, 8H), 3.88 (m, 16H), 3.64 (dd, *J* = 4.0 Hz, *J* = 7.6 Hz, 4H), 2.95–2.50 (m, 8H), 2.40–2.30 (m, 8H), 1.35–1.20 (m, 72H), 0.84 (m, 48 H). ^{13}C{^{1}H} NMR (100 MHz, DMSO-*d*$_6$): δ 171.06, 168.36, 147.92, 145.06, 136.54, 130.07, 126.99, 66.25, 66.18, 63.26, 61.50, 38.19, 38.14, 18.12, 34.14, 29.75, 29.64, 29.58, 28.36, 23.22, 23.19, 23.03, 22.44, 22.41, 13.95, 13.92, 10.85, 10.82, 10.78. Anal. calcd for $C_{116}H_{180}N_4O_{28}S_4$: C, 63.13; H, 8.22; N, 2.54%; found: C, 60.09; H, 8.22; N, 2.54%.

Cyclobis(paraquat-p-phenylene) Tetrakis[Di(2-butyloctyl) Sulfosuccinate] (1·3). This compound was prepared from **12** (66 mg, 0.1 mmol) and **Na·3** (278 mg, 0.5 mmol) in a similar manner to **1·2**. The formed precipitate was collected by decantation, washed with water, and recrystallized from acetone/water to give **1·3** as a white waxy solid (138 mg, 52%). ^{1}H NMR (400 MHz, DMSO-*d*$_6$): δ 9.46 (d, *J* = 6.4 Hz, 8H), 8.66 (d, *J* = 6.4 Hz, 8H), 7.17 (s, 8H), 5.81 (s, 8H), 3.86 (m, 16H), 3.62 (dd, *J* = 4.0 Hz, *J* = 7.6 Hz, 4H), 2.95–2.50 (m, 8H), 2.40–2.30 (m, 8H), 1.35–1.20 (m, 128H), 0.84 (m, 48 H). ^{13}C{^{1}H} NMR (100 MHz, DMSO-*d*$_6$): δ 171.02, 168.29, 147.90, 145.05, 136.53, 130.07, 126.97, 66.57, 66.48, 63.28, 61.51, 36.70, 36.63, 34.15, 31.28, 31.23, 30.58, 30.48, 30.28, 30.11, 29.05, 29.00, 28.34, 26.06, 22.44, 22.41, 22.13, 22.09, 13.96, 13.91. Anal. calcd for $C_{148}H_{244}N_4O_{28}S_4$: C, 66.93; H, 9.26; N, 2.11%; found: C, 65.99; H, 9.32; N, 2.08%.

Preparation for 1·2/AOT (1:1). To a methanol solution of **1·2** (4.41 mg, 0.002 mmol) was added a methanol solution of AOT (0.889 mg, 0.002 mmol). The solvent was evaporated and the residue was dried under vacuum. Other samples (**1·2**/AOT (1:2) and **1·2**/AOT (1:4)) are similarly prepared.

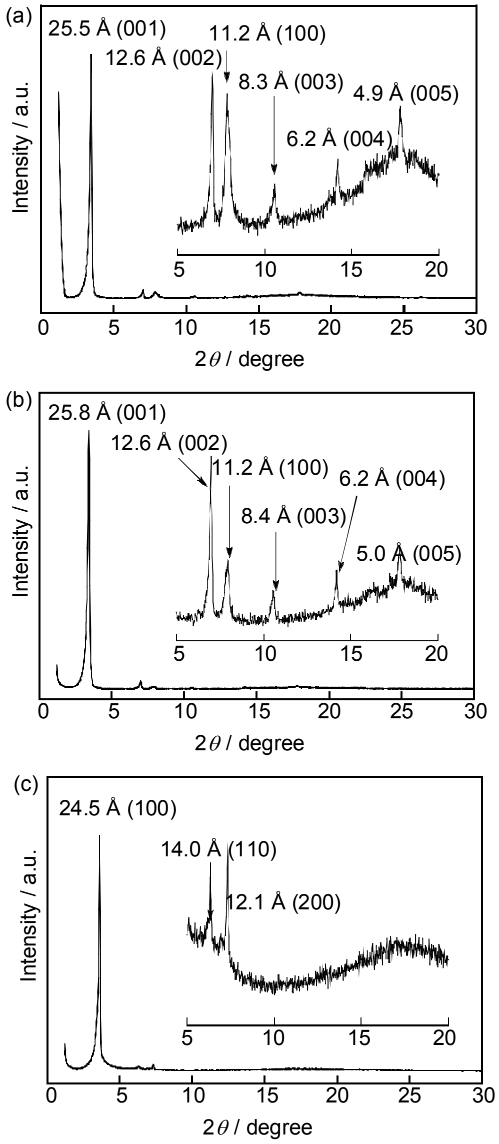


Fig. S2 X-ray diffraction patterns of (a) **1·2**/AOT (1:0.5) in the L_{Col} phase at 200 °C, (b) **1·2**/AOT (1:1) in the L_{Col} phase at 200 °C, and (c) **1·2**/AOT (1:4) in the Col_h phase at 200 °C.

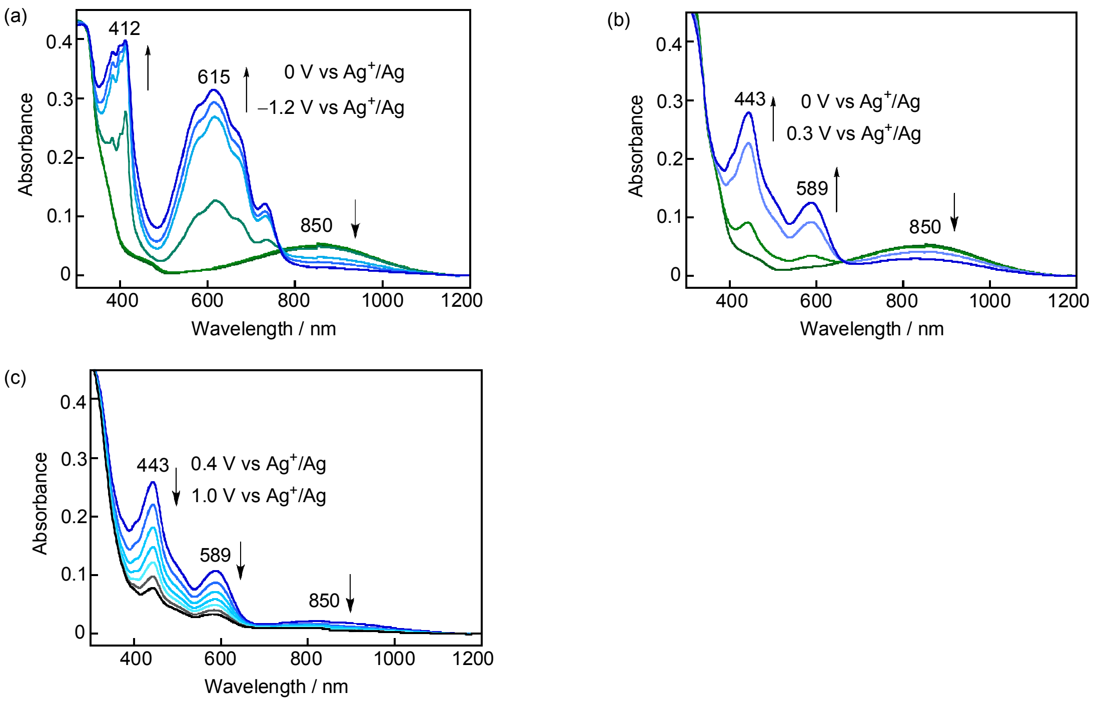


Fig. S3 UV–vis–NIR spectra of **1·2**/TTF (1:1) in a DMSO solution during (a) the reduction, (b) the first oxidation, and (c) the second oxidation. These spectra are recorded in DMSO solutions of Bu_4NClO_4 (0.10 M) with Pt working electrodes. The sweep rate is 100 mV s^{-1}. Arrows indicate the direction of spectral changes.

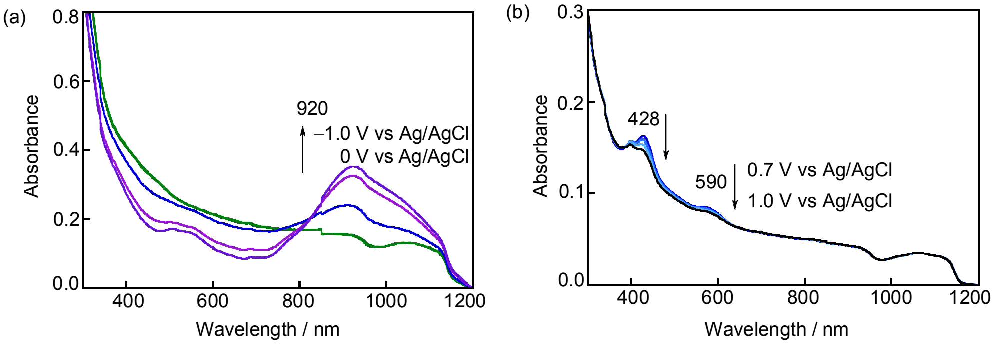


Fig. S4 UV–vis–NIR spectra of **1·2**/TTF (1:1) in a thin film during (a) the reduction and (b) the second oxidation. These spectra are recorded in aqueous solutions of LiCl (0.10 M) with ITO working electrodes. The sweep rate is 30 mV s^{-1}. Arrows indicate the direction of spectral changes.

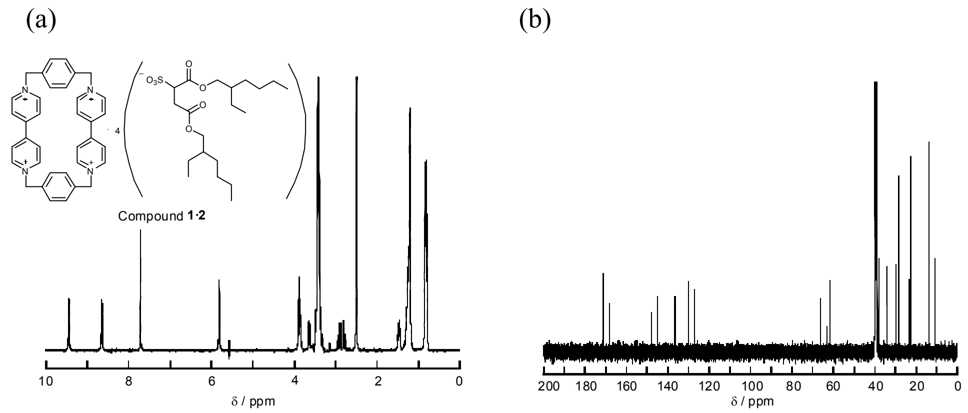


Fig. S5 (a) ^{1}H NMR and (b) ^{13}C NMR spectra of **1·2** in DMSO-d_6.

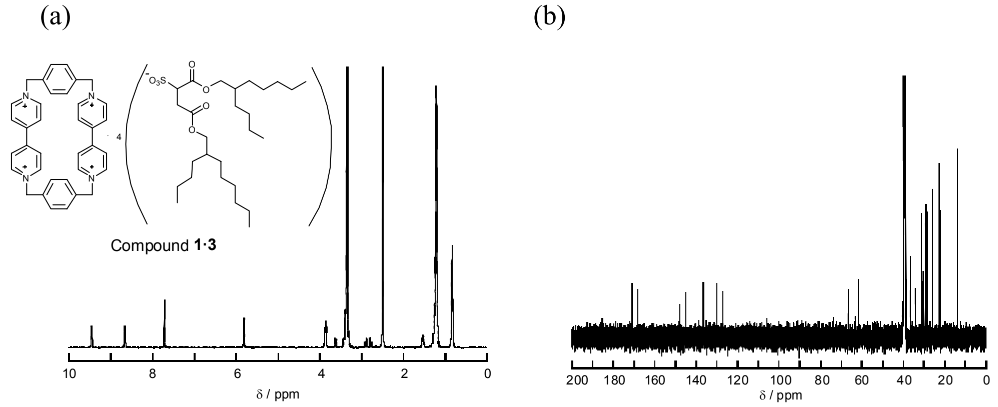


Fig. S6 (a) ^{1}H NMR and (b) ^{13}C NMR spectra of **1·3** in DMSO-d_6.

Reference.

[1] B. Odell, M. V. Reddington, A. M. Z. Slawin, N. Spencer, J. F. Stoddart and D. J. Williams, *Angew. Chem., Int. Ed. Engl.*, 1988, **27**, 1547.