Electronic Supplementary information
Conversion of curved assemblies into two dimensional sheets

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Experimental Section:

Chemicals and materials: Unless otherwise stated, all the chemicals and reagents were obtained commercially and used without further purification. 1,3-dibromopropane, 1,4-dibromobutane, 1,5-dibromopentane, 16-bromohexadecane, sodium azide, 11-Bromoundecanoic acid, 2-Ethyl-hexylamine and Ethylchloroformate were purchased from Sigma Aldrich and used as received. Indoline-2,3-dione and indolin-2-one were purchased from Spectrochem Chemicals. All the solvents were purchased from Merk Chemicals. All the solvent used for self-assembly studied were HPLC grade.

General Methods and Instruments: Analytical Thin Layer Chromatography was done on pre-coated silica gel plates (Kieselgel 60F254, Merck). Column chromatographic purifications were done with 60-120 mesh sized silica gel. All the $^1$H NMR spectra were recorded in CDCl$_3$ on Bruker arx AV 200 MHz and AV 400 MHz Bruker AVANS spectrometer. High temperature $^1$H NMR spectra were recorded in 1,1,2,2 tetrachloroethane-d$_2$ on Bruker arx AV 400 MHz. $^{13}$C NMR spectra were measured on Bruker arx 100 MHz AVANS spectrometer. All chemical shifts are reported in $\delta$ ppm downfield to TMS and peak multiplicities are referred as singlet (s), doublet (d), triplet (t), quartet (q), pentet (p), and multiplet (m). UV-vis absorption spectra were recorded on SPECORD® 210 / PLUS, UV-visible spectrophotometer. Variable temperature UV-vis spectra were record on Agilent 8453 UV-visible Spectroscopy System connected with Agilant 89090A temperature controller. Emission spectra were recorded on FLUOROMAX-4 C research spectrofluorometer. MALDI-TOF analysis was done on Voyager-De-STR MALDI-TOF (Applied Bio systems, Framingham, MA, USA) equipped with 337-nm pulsed nitrogen laser used for desorption and ionization, 1 $\mu$M solution of sample was premixed with DHB (2, 5-dihydroxy benzoic acid) matrix in CHCl3 and mixed well before spotting on 96-well stainless steel MALDI plate by dried droplet method for MALDI analysis.
TEM imaging was done with a Jeol 1200 EX transmission electron microscope. The samples were prepared by drop casting (5-10 µl of 5×10⁻⁴ M) of the sample on the carbon coated copper grids (400 grids) obtained from Ted Pella. The samples were dried at 50 °C for 12 h before analysis. For Ageing studies, Drop casted samples were stored under normal environment for 20 days and then analysed by TEM. AFM measurement was done by drop casting (5-10 µl of 5×10⁻⁴ M) of sample on ZP-P4VP spun silicon wafers. DFT studies were performed using the B3LYP functional and polarized 6-31g* basis set for the geometry optimization.

**General synthetic route:**

![Scheme S1](image)

**Scheme S1.** Synthesis of target molecules

**Procedure:**

**N-(2-ethylhexyl) N’(10-bromodecyl) urea (C):** In 200 mL round bottom flask equipped with magnetic stirrer, 11-bromoundecanoic acid (a) (5 g; 18.8 mmol) and triethylamine (2.90 mL; 22.6
mmol) was dissolved in dry THF at 0°C under argon atmosphere. To the mixture, ethylechlororformate (2.44 mL; 22.6 mmol) was added drop wise over a period of 15-20 minutes and stirred at 0°C for 3 h. Sodium azide (1.4 g; 22.56 mmol) dissolved in cold DI water was added to the mixture and stirred at 0°C followed by another hour at room temperature. Completion of reaction was monitored by thin layer chromatography using hexane as a mobile phase. Reaction mixture was poured into ice-cold water and washed twice with brine solution. The combine organic phase was extracted with ethyl acetate and dried over Na$_2$SO$_4$. Excess of solvent was removed under reduced pressure at room temperature water bath and pass through short pad silica column to get 11-bromoundecanoiyl azide as a pale yellow oil (5 g, yield = 91 %). The 11-bromoundecanoiyl azide was dissolved in Dry THF and refluxed for overnight to get Isocynate b, the reaction was cooled to room temperature and 2-ethylhexyl amine (3.26 mL; 18.86 mmol) dissolved in dry CHCl$_3$ and cat. Triethylamine was added to it. The reaction mixture was then stirred at room temperature for 12 h. Reaction mixture was washed with water and brine solution and extracted with dichloromethane. Excess of solvent was removed under reduced pressure. The pale yellow crude oil was purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (2:8) as mobile phase to get final compound C (4.6 g, yield= 61 %). $^1$H-NMR (200MHz, CDCl$_3$): $\delta$ 4.41 (broad s, 2H, Urea N-H), 3.40 (t, 2H, J=6.8 Hz) 3.16 (t, 2H, J=6.2 Hz), 3.13 (m, 2H) 1.84 (p, 1H, J= 7 Hz), 1.36 (m, 24H), 0.88 (t, 6H, J=7.2 Hz). $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 159.29 (urea carbonyl), 43.9, 41.1, 40.4, 33.3, 31.5, 30.04, 29.9, 24.7, 23.6, 14.6, 11.4; HR-LCMS (ESI) m/z calculated for [M+H]+: 391.4, found 391.23

1,3 bis(Indoline1,3dione)propane D1: 100 mL round bottom flask equipped with magnetic stirrer was charged with Indoline2,3-dione (1 g, 6.80 mmol) and K$_2$CO$_3$ (1.87 g, 13.60 mmol) in DMF. The resultant mixture was heated at 80 °C for half hour followed by drop wise addition of 1,3-dibromopropane (0.345 mL, 3.4 mmol) under Argon atmosphere. The temperature was raised up to 100 °C and heated for 18 h. Completion of reaction was monitored by TLC. The reaction mixture was cooled down to room temperature. 250 mL ethyl acetate was added to it, combined organic phase was washed twice with water and brine solution. The organic phase was dried over Na$_2$SO$_4$. Excess of solvent was removed under reduced pressure and crude product was purified by column
chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (4:6) as mobile phase to get yellow solid product 1,3 bis(Indoline1,3dione)propane D1. (600 mg, Yield= 53 %). ^1H-NMR (200 MHz; CDCl$_3$) $\delta$ 7.59 (m, 4H), 7.14 (t, 2H, J=7.8 Hz), 6.9 (d, 2H, J = 8.3 Hz), 3.87 (t, 4H, J=7.1 Hz), 2.23 (m, 2H, -CH$_2$). ^13C-NMR (100 MHz; CDCl$_3$) $\delta$ 182.7(carbonyl), 158.3(amide carbonyl), 150.1, 138.5, 125.6, 124, 117.5, 109.9, 38.1, 25.1; HR-LCMS (ESI) m/z calculated for [M+Na]$^+$: 348.10, found 357.09.

1, 4 bis (Indoline1,3 dione)butane D2: synthesized by using similar procedure used for compound D1. Indoline2,3-dione (1 g, 6.80 mmol), K$_2$CO$_3$ (1.87 g, 13.60 mmol and 1,4-dibromobutane (0.4 mL, 3.4 mmol) purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (4:6) as mobile phase to get yellow solid product. (640 mg, Yield= 54 %). ^1H-NMR (200 MHz; CDCl$_3$) $\delta$ 7.6 (m, 4H), 7.13 (t, 2H, J=7.4 Hz), 6.93 (d, 2H, J=8 Hz), 3.81(t, 4H, J=6 Hz) 1.83 (m, 4H). ^13C-NMR (100 MHz; CDCl$_3$) $\delta$ 182.7(carbonyl), 158.4(amide carbonyl), 150.4, 138.5, 125.5, 123.9, 117.5, 110.1, 48.9, 39.4, 24.4; HR-LCMS (ESI) m/z value calculated for C$_{20}$H$_{26}$O$_4$N$_2$ [M+H]$^+$: 348.9, found 349.02

1, 5 bis (Indoline1,3 dione)pentane D3: synthesized by using similar procedure used for compound D1. Indoline2,3-dione (1 g, 6.80 mmol), K$_2$CO$_3$ (1.87 g, 13.60 mmol and 1,5-dibromopentane (0.460 mL, 3.4 mmol). Purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (3:7) as mobile phase to get yellow solid product (700 mg, Yield=57 %). ^1H-NMR (200 MHz; CDCl$_3$) $\delta$ 7.61 (m, 4H), 7.13 (d, 2H, J=7.4 Hz), 6.93 (d, 2H, J= 8.2 Hz), 3.74 (t, 4H J=7Hz), 1.83 (m, 4H), 1.47 (m, 2H). ^13C-NMR (100 MHz; CDCl$_3$) $\delta$ 182.78(carbonyl), 158.6 (amide carbonyl), 150.1, 137.84, 124.85, 123.12, 116.89, 109.51, 39.08, 25.95, 23; HR-LCMS (ESI): m/z value calculated for C$_{21}$H$_{28}$O$_4$N$_2$ [M+Na]$^+$: 384.98, found 385.11

1, 3 bis(Isoindigo)propane E1: In second step, 1, 3 bis(Indoline2,3-dione)Propane D1 (500 mg, 0.890 mmol) and oxindole (258 mg, 1.95 mmol) were dissolved in mixture of 20 mL glacial acetic acid and catalytic amount of concentrated HCl (2-3 drops). The reaction mixture was heated to 90 ºC for 12 h. After completion it was cooled down to room temperature and 200 mL Dichloromethane was
added. The resultant mixture was washed with water and organic layer was separated. The separated organic layer was dried over Na$_2$SO$_4$ and excess of solvent was removed under reduced pressure. Crude product was purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (4:6) as mobile phase to get red colour solid. (480 mg Yield= 57 %). $^1$H-NMR (200 MHz; CDCl$_3$) δ 9.1(d, 4H, J=8.1 Hz), 7.82 (s, 2H), 7.51 (m, 2H), 7.35 (dd, 4H, J=3.1, 1.1 Hz), 7.07 (t, 2H, J= 7.8 Hz), 6.8 (m, 2H), 3.91 (m, 4H), 2.17 (m, 2H). $^{13}$C-NMR could not be recorded because of low solubility. Maldi-Tof m/z value calculated for C$_{33}$H$_{24}$O$_4$N$_4$ [M+Na]$^+$: 587.18, found 586.96

1, 4 bis(Isoindigo)butane E2: synthesized by using similar procedure used for compound E1. D2 (550 mg, 0.953 mmol) and oxindole (277 mg, 2.09 mmol). Purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (3:7) as mobile phase to get red colour solid. (590 mg, Yield= 64%). $^1$H-NMR (200 MHz; CDCl$_3$) δ 9.1(dd, 4H, J=7.5 Hz), 7.7 (s, 2H), 7.5 (dd, 2H, J=7.5 Hz), 7.34 (m, 2H), 7(m, 4H), 6.8 (m, 4H), 3.91 (m, 4H), 2.17 (m, 4H). $^{13}$C-NMR could not be recorded because of low solubility. Maldi-Tof m/z value calculated for C$_{36}$H$_{26}$O$_4$N$_4$ [M+H]$^+$: 579.20, found 579.20

1, 5 bis(Isoindigo)pentane E3 or 4: synthesized by using similar procedure used for compound E1. D3 (600 mg, 1.01 mmol) and oxindole (294 mg, 2.22 mmol). Purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (3:7) as mobile phase to get red colour solid. (640 mg, Yield= 65 %). $^1$H-NMR (200 MHz; CDCl$_3$) δ 9.13 (t, 4H, J = 8.2 Hz), 7.85 (s, 2H), 7.5 (m, 2H), 7.3 (d, 2H), 7.08(m, 4H), 6.9 (d, 2H, J=7.9 Hz), 6.8 (m, 2H). $^{13}$C-NMR (100 MHz; CDCl$_3$) δ 169.8, 151.1, 145.1, 143, 138.9, 133.2, 130.5, 124, 122.2, 110.7, 108.5, 40.5, 24.7; HR-LCMS ESI: m/z value calculated for C$_{37}$H$_{28}$O$_4$N$_4$ [M+H]$^+$: 592.21, found 593.01

General synthetic procedure for the synthesis of 1, 2 and 3:

In third step, respective compound (E1 or E2 or E3) and K$_2$CO$_3$ were dissolved in DMF at 80 °C for half hour in argon atmosphere. Compound C dissolved in DMF added drop wise to the reaction mixture and heated at 90 °C for 14 h under argon atmosphere. Completion of reaction was monitored
by TLC. After completion, reaction was cooled to room temperature and 200 mL of ethyl acetate was added. Combined organic layer was washed with water and brine solution. The organic layer was separated and dried over Na$_2$SO$_4$. The excess of solvent was removed under reduced pressure. Crude product was purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (2:8) as mobile phase to get respective compound 1, 2 and 3 respectively.

**Compound 1:**

Compound E1 (300 mg, 0.531 mmol), K$_2$CO$_3$ (293 mg, 2.12 mmol) and C (500 mg, 1.327 mmol).

Purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (2:8) to get red color solid (355 mg, Yield = 56 %).$^1$H-NMR (200 MHz; CDCl$_3$) $\delta$ 9.13 (dd, 4H, J= 7.9, 3.9 Hz), 7.45 (dd, 4H, J= 7.9, 2.9 Hz), 7.35 (m, 4H), 7.05 (t, 2H, J= 7.3 Hz), 6.78 (m, 2H), 4.47 (br, s, 4H), 3.84 (m, 8H), 3.11 (m, 8H), 2.19 (m, 2H), 1.74 (m, 2H), 1.3 (br, s, 44H), 0.87 (t, 12H, J = 6.2 Hz). $^{13}$C-NMR (100 MHz; CDCl$_3$) $\delta$ 167 (d, J= 33.1 Hz), 158 (d, J = 30.8 Hz), 150, 144.7, 138.7, 133.9, 132 (d, J = 23.1 Hz), 129, 125, 123.4, 122.5, 117.5, 109.9, 107.9 (d, J= 30 Hz), 43.4, 37.6, 30.9, 30.1, 29.1, 27.3, 26.9, 25.2, 24.1, 23, 14, 10. HR-LCMS ESI: m/z value calculated for C$_{73}$H$_{100}$O$_6$N$_8$ [M]+: 1184, found 1184.67

**Compound 2:**

Compound E2 (300 mg, 0.519 mmol), K$_2$CO$_3$ (286 mg, 2.07 mmol) and C (487 mg, 1.29 mmol).

Purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (2:8) to get red color solid (370 mg, yield = 60 %). $^1$H-NMR (200 MHz; CDCl$_3$) $\delta$ 9.13 (dd, 4H, J= 7.9, 3.9 Hz), 7.57 (dd, 4H, J= 6.7, 1 Hz), 7.39 (m, 4H), 7.05 (td, 2H J= 6, 6.19, 1.77 Hz), 6.8 (m, 2H), 4.58 (br s, 2H), 4.44 (br s, 2H), 3.79 (m, 8H), 3.11 (m, 8H), 1.75 (m, 6H), 1.3 (br s, 44H), 0.87 (t, 12H, J= 6.1 Hz). $^{13}$C-NMR (100 MHz; CDCl$_3$) $\delta$ 167 (d, J= 33.1 Hz), 158.4, 150.8, 144.7, 144.3, 133.6, 132.4, 129.8, 125.4, 123.6, 122.3, 121.6, 117.5, 110.1, 107.9, 43.3, 40.5, 39.9,
30.1, 29.1, 28.6, 27.3, 26.8, 24.1, 23, 14, 10. HR-LCMS ESI: m/z value calculated for C_{74}H_{102}O_{6}N_{8} [M]+: 1198.79

**Compound 3:**

Compound E3 (300 mg, 0.507 mmol), K_{2}CO_{3} (280 mg, 2.02 mmol) and C (476 mg, 1.267 mmol). Purified by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (2:8) to get red colour solid (380 mg, yield=62 %) ¹H-NMR (200 MHz; CDCl₃) δ 9.13(dd, 4H, J=7.9, 3.9 Hz), 7.55 (dd, 4H, J=7.7, 2.9 Hz), 7.32 (t, 4H, J=7.3 Hz), 7.05 (q, 2H J= 7.9 Hz), 6.8 (m, 2H), 4.65 (br s, 4H), 3.73 (m, 8 H), 3.09 (m, 8H), 1.73 (m, 6H), 1.39 (br. S, 46H), 0.87 (t, 12H, J= 5.8 Hz). ¹³C-NMR (100 MHz; CDCl₃) δ 167, 158, 150, 144, 138, 133 (d, J=21.5 Hz), 129, 125, 123, 122, 117, 110, 107, 43, 39, 30 (d, 38.06 Hz), 29 (d, J= 15.73), 27 (d, J= 24.88 Hz), 24, 23, 14. HR-LCMS ESI m/z value calculated for C_{75}H_{104}O_{6}N_{8} [M+Na]+: 1235.79, found 1235.79

**Synthetic procedure for 5:** 100 mL round bottom flask equipped with magnetic stirrer was charged with Compound E3 (200 mg, 0.338 mmol) and K₂CO₃ (184 mg, 1.35 mmol) were dissolved in DMF at 80 °C for half hour under argon atmosphere. 1-bromohexadecane (257 µL, 0.845 mmol) was added drop wise to the reaction mixture and heated at 90 °C for 14 h under argon atmosphere. Completion of reaction was monitored by TLC. After completion, reaction was cooled to room temperature and 200 mL of ethyl acetate was added. Combined organic layer was washed with water and brine solution. The organic layer was separated and dried over Na₂SO₄. The excess of solvent was removed under reduced pressure. crude product was purifed by column chromatography using 60-120 mesh sized silica gel as stationary phase and ethyl acetate/Pet ether (1:19) as mobile phase to get shiny red colour solid. (250 mg, Yield=62 %). ¹H-NMR (200 MHz; CDCl₃) δ 9.1 (dd, 4H, J=8, and 4.1 Hz), 7.56 (m, 2H), 7.37 (m, 2H), 7.08 (m, 4H), 6.86 (d, 2H, J=8 Hz), 6.81 (d, 2H, J=7.8 Hz), 3.74 (dt, 4H, J= 6.65 and 7.1 Hz), 1.71 (m, 6H), 1.34 (br s, 22H), 0.88(t, 6H, J=6 Hz). ¹³C-NMR (100 MHz; CDCl₃) δ 167.7, 158.1, 150.8, 144.7, 138.3, 132.4, 129.9, 125.4, 122.3, 121.6, 117.5, 110.1, 107.8, 39.9, 31.9, 29.3, 27.4, 26.9, 24.1, 22.6, 14.1; HR-LCMS m/z value calculated for C_{69}H_{92}O_{4}N_{4} [M+H]+: 1041.70, found 1041.30.
**Synthetic procedure for 6:** In a 50 mL round bottom flask equipped with magnetic stirrer Compound 3 (100 mg, 0.00809 mmol) and activated Zn dust (32 mg 0.0485 mmol) were added to Dry THF (10 mL) and cooled to 0 °C. TFA (7 μL 0.485 mmol) was added dropwise. The mixture was stirred at r.t. for 1 hour. Then the mixture was added to 20 mL Chloroform the combined organic phase was washed with H₂O (50 mL). The organic layer was separated and dried over Na₂SO₄. The excess of solvent was removed under reduced pressure. The final compound was collected as a yellowish liquid. (70 mg, yield = 70 %).

Disappearance of peak at δ 9.1 ppm (which is characteristic for the i-Indigo) confirms the formation of product. We were not able to record the 13C-NMR because of low solubility.

1H-NMR (200 MHz; CDCl₃) 7.63-7.49 (m, 2H), 7.28-7.07 (m, 4H), 6.86-6.97 (m, 2H), 5.35 (br S, 2H), 4.11 (d, 2H), 3.74- 3.49 ( m, 8 H,), 2.99-3.06 (m, 8H), 1.34 (br s, 22H), 0.88(t, 6H).
Figure S1: Concentration dependent $^1$H-NMR for 2 (a) and 3 (b) (Solvent: CDCl$_3$) and Temperature dependent $^1$H-NMR for 1 (c), 2 (d) and 3 (e); Solvent: 1,1,2,2-Tetrachloroethane-d$_2$ (Red: Room Temperature and Green: at 60 °C)
**Figure S2:** UV-vis absorption spectrum of $5 \times 10^{-5}$ M of 2 (a), 3 (b), 4 (c), 5 (d) in different solvent and 2 (e), 3 (f) in the mixture of THF and Chloroform.

**Figure S3:** Variable Temperature UV-vis absorption of 1 (a), 2 (b) and 3 (c) at different temperature

**Figure S4:** Variable Temperature UV-vis absorption plot ($I_{391}/I_{369}$ vs $T$) of 1 (a), 2 (b) and 3 (c) at different temperature
To obtain toroidal morphology, the molecules were dissolved in dry chloroform (0.5 mM) and the solution was kept quiescent for 12 h. Then, the solution was drop casted on the TEM grid and dried at room temperature for 12 h before imaging. For AFM, the solution was drop casted on silicon wafer and dried at room temperature for 12 h. To obtain Sheet morphology, the molecules were dissolved in Dry THF and drop casted on TEM grid (for TEM) and Silicon wafers (for AFM). The samples were dried at room temperature before analysis.

**Figure S5:** TEM image of 2 prepared from chloroform

**Figure S6:** AFM image and height profile of molecule 2

**Figure S7:** Fibre protruding from toroid
Figure S8: Transmission Electron microscopy (TEM) images of 2 (a-d) and molecule 3 (e-h) after 20 days of drying at room temperature; (c = 5x10^{-4} M; Solvent: CHCl_3)

Figure S9: DFT Energy Minimize structure of 3

Figure S10: TEM image of 1 in THF
Figure S11: TEM image of 2 in THF

Figure S12: AFM image of 2 in THF and water mixture

Figure S13: UV-vis absorption spectrum of 5 x 10^{-5} M of 4 in different solvent.
Figure S14: TEM image of 4 in THF

Figure S15: UV-vis absorption spectrum of 5 x 10^{-5} M Solution of 5 in different solvent.

Figure S16: UV-vis absorption spectrum of 5 x 10^{-5} M solution of 6 in THF.
Figure S17: UV-vis absorption spectrum of $5 \times 10^{-5}$ M solution of 6 in Chloroform.

Further we have reduce the aggregation in NMR experiments by adding the H-bond acceptor solvent such as methanol (Figure S18). In presence of methanol the peaks corresponding to urea moiety disappeared (red line in Figure S18).

Figure S18: $^1$H-NMR aggregation experiment with CDCl$_3$ (Blue) and CDCl$_3$:10% Methanol-d$_4$ (Red)
Figure S19: H type aggregates are due to π-stacking in absence of hydrogen bonding (a) and J type aggregate due to the presence of π-stacking as well as hydrogen bonding.

Figure S20: Concentration dependent $^1$H-NMR of 1 with higher concentration (a), with lower concentration (b).
$^1$H-NMR of 1:

![H-NMR spectrum]

$^{13}$C-NMR of 1:

![C-NMR spectrum]
$^{1}H$-NMR of 2:

$^{13}C$-NMR of 2:
$^1$H-NMR of 3:

$^{13}$C-NMR of 3:
$^1$H-NMR of 4:

$^{13}$C-NMR of 4:
$^1$H-NMR of 5:

$^{13}$C-NMR of 5:
$^1$H-NMR of 6: