

**SUPPLEMENTARY INFORMATION**

**Light-induced twisting, untwisting, and retwisting of aromatic polyamides:  
An interplay between the induced chirality and the co-facial  $\pi$ -stacking  
interactions**

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## Table of contents

Contents	Page no.
1. Materials .....	2
2. Methods .....	2
3. Synthetic scheme for the preparation of Phe(L or D or D/L)-N <sub>3</sub> .....	4
4. Synthetic scheme for the preparation of precursor polymer and azido acid functionalized aromatic polyamides .....	5
5. Synthesis of model compounds .....	13
6. Concentration-dependent UV-Vis and CD studies.....	17
7. Helicity induction and single handedness.....	17
8. TEM Imaging .....	19
9. Effect of the guest molecule on the helical folding .....	20
10. Twisting – Untwisting - Retwisting of aromatic polyamides.....	27
11. References .....	30

## Experimental procedure

### 1. Materials

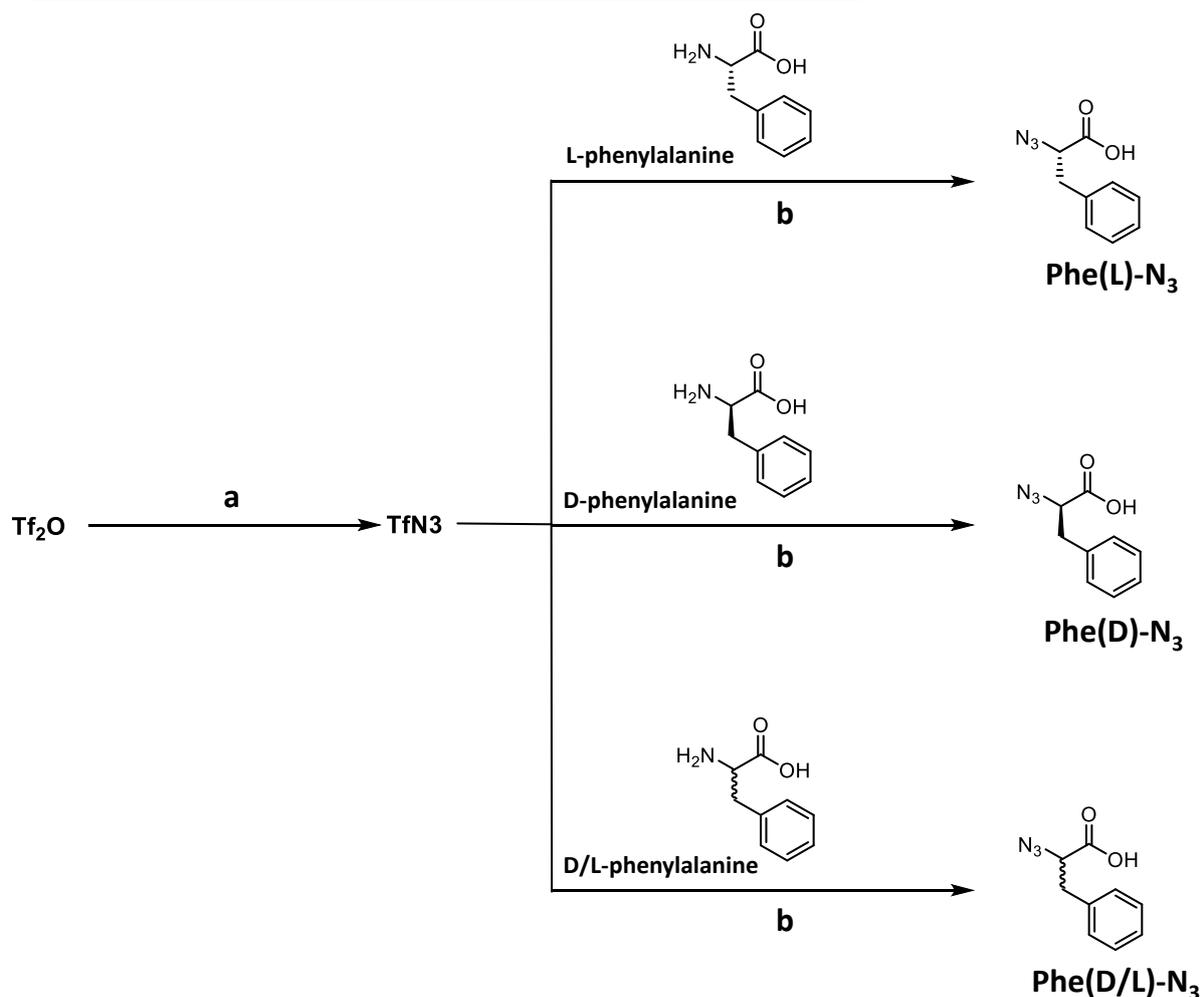
Chelidamic acid, pyrene, propargyl alcohol, Triflic anhydride, L or D or D/L-phenylalanine, 1-Ethyl-3-(3-dimethylaminopropyl) carbodiimide (EDC), 4-dimethylaminopyridine (DMAP), Tosyl chloride, NaSH, hexamethylphosphoramide (HMPA), N-Methyl-2-pyrrolidone (NMP), spiropyran (Sp), all the deuterated solvents, etc. were purchased from Sigma Aldrich. KOH, NaOH, Oxalyl chloride, sodium azide, dimethyl sulfoxide (DMSO), CuSO<sub>4</sub>, Na-ascorbate, K<sub>2</sub>CO<sub>3</sub>, triethylamine (TEA), trifluoroacetic acid (TFA), etc., and all the common organic solvents were purchased from both TCI Chemicals and Avra and were used without further purification. Solvents like DCM, methanol, THF, HMPA, and NMP were dried using standard drying procedures<sup>1</sup> and distilled using a standard distillation setup, while HMPA and NMP were distilled using the Kugelrohr apparatus. Purification of products was carried out by column chromatography using silica gel of mesh size of 100-200 μm and 60-120 μm. Thin-layer chromatography (TLC) was carried out on pre-coated plates. UV light (254 nm) and ninhydrin stain were used to visualize nitrogen-containing compounds.

### 2. Methods

The <sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded on Bruker Biospin Avance III FT-NMR 400 MHz spectrometer, with tetramethylsilane (TMS) as an internal standard at room temperature. NOESY spectra was recorded on Bruker Biospin Avance III FT-NMR 600 MHz spectrometer, with tetramethylsilane (TMS) as an internal standard at room temperature. NMR data were processed using Mestre Nova software. UV-Vis spectral measurements were carried out with

Agilent Cary 60 UV-Vis spectrophotometer using 1 cm path length quartz cuvettes. The steady-state emission studies were performed using FL Solution software with the Hitachi F7000 fluorescence spectrophotometer. GPC measurements were carried out with a Malvern Omnisec instrument having a refractive index (RI) detector using Shodex GPC KD-806M column with DMF with 0.01 M LiBr as eluent at 35 °C with a flow rate of 0.7 mL/min and PMMA as standard for all the samples. The results were analyzed by using Omnisec software. The sample peaks were analyzed for  $M_n$ ,  $M_w$ , and  $\bar{M}$  using the conventional calibration. The Circular dichroism studies were performed using a Chirascan VX Applied Photophysics spectrophotometer. Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) images were recorded using JEOL JEM-F200 equipped with electron-dispersive X-ray spectroscopy (EDS). Hydrodynamic radii were estimated using dynamic light scattering (DLS) measurements on a Malvern Zetasizer Nano ZS90 instrument. All the data were plotted using Origin 2018 software. Molecular structures and reaction schemes were drawn using ChemDraw Professional 15.0 software.

### 3. Synthetic scheme for the preparation of Phe(L or D or D/L)-N<sub>3</sub>



(a) = NaN<sub>3</sub>, H<sub>2</sub>O, DCM, 4 h., 0 °C

(b) = K<sub>2</sub>CO<sub>3</sub>, CuSO<sub>4</sub>·5H<sub>2</sub>O, H<sub>2</sub>O, CH<sub>3</sub>OH, RT, 12 h.

Scheme S1. Synthetic scheme of azido acids (Phe(L or D or D/L)-N<sub>3</sub>).

#### Triflic azide (TfN<sub>3</sub>)<sup>2</sup>

To sodium azide (3.9 gm, 59.52 mmol) mixture of H<sub>2</sub>O (9.7 ml) and DCM (16.3 ml) was added, and the resulting solution was cooled to 0 °C in an ice bath. Triflic anhydride (Tf<sub>2</sub>O) (2.027 ml, 12.09 mmol) was added into the reaction flask slowly over 5 mins, and the reaction was stirred vigorously at 0 °C for 4 hours. After completion of the reaction, the organic layer was separated, and the aqueous layer was extracted with DCM (2 × 8.2 ml). The combined organic layer containing triflyl azide (TfN<sub>3</sub>) was washed with saturated Na<sub>2</sub>CO<sub>3</sub> solution (8.2 ml) and used for the next step without further purification.

#### Azido acids (Phe(L or D or D/L)- N<sub>3</sub>)<sup>2</sup>

To a round bottom flask, amino acid (L or D or D/L-phenylalanine) (1 gm, 6.05 mmol) was combined with K<sub>2</sub>CO<sub>3</sub> (1.26 gm, 9.124 mmol), CuSO<sub>4</sub>·5H<sub>2</sub>O (15.1 mg, 0.0605 mmol), H<sub>2</sub>O (20

ml) and CH<sub>3</sub>OH (40 ml). TfN<sub>3</sub> in DCM solution was added to this reaction mixture and the resulting solution was stirred at room temperature for 14 hours. After completion, the organic solvent was evaporated under reduced pressure and the aqueous layer was adjusted to pH = 6 using 2 M HCl. The aqueous solution was extracted with ethyl acetate (3 × 20 ml) to remove any by-products. The aqueous solution was further acidified to pH = 2 using 2 M HCl. The resulting precipitates were extracted by ethyl acetate (3 × 20 ml) and the organic extracts were combined, passed through Na<sub>2</sub>SO<sub>4</sub>, and evaporated to dryness to get the desired product as a colorless oil.

Phe(L)-N<sub>3</sub> (yield: 67 %)

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 13.43 (s, 1H, -COOH), 7.37 – 7.21 (m, 5H, Ar-H), 4.39 (dd, 1H, -CHN<sub>3</sub>CHH'-), 3.11 (dd, 1H, -CHH'Ar), 2.92 (dd, 1H, -CHH'Ar).

[α]<sub>D</sub><sup>25</sup> = -51.92 (C = 0.12 gm/100 ml in CHCl<sub>3</sub>).

Phe(D)-N<sub>3</sub> (yield: 73 %)

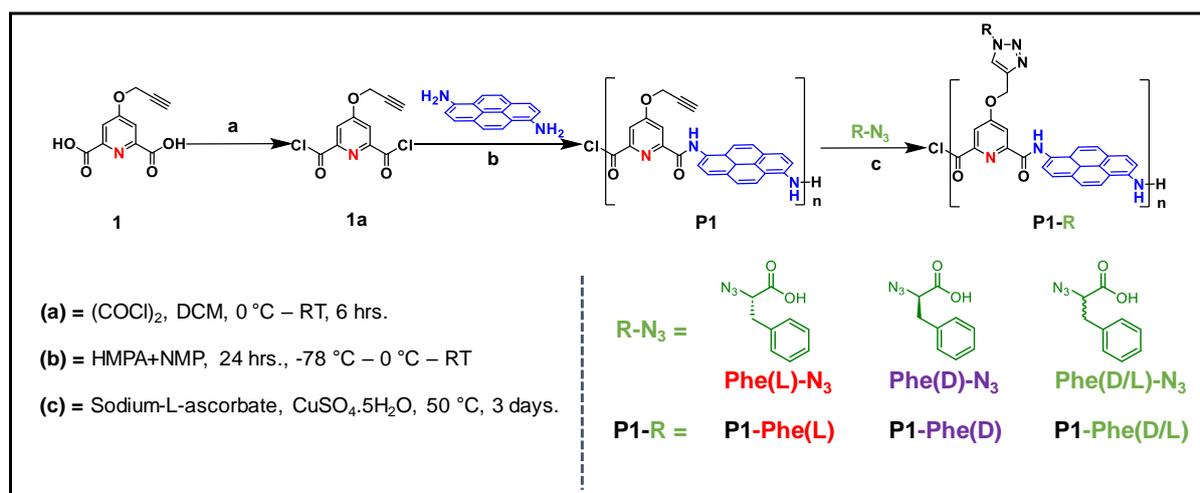
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 13.40 (s, 1H, -COOH), 7.40 – 7.20 (m, 5H, Ar-H), 4.36 (dd, 1H, -CHN<sub>3</sub>CHH'-), 3.12 (dd, 1H, -CHH'Ar), 2.90 (dd, 1H, -CHH'Ar).

[α]<sub>D</sub><sup>25</sup> = +52.68 (C = 0.099 gm/100 ml in CHCl<sub>3</sub>).

Phe(D/L)-N<sub>3</sub> (yield: 70 %)

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 13.41 (s, 1H, -COOH), 7.37 – 7.18 (m, 5H, Ar-H), 4.37 (dd, 1H, -CHN<sub>3</sub>CHH'-), 3.12 (dd, 1H, -CHH'Ar), 2.91 (dd, 1H, -CHH'Ar).

#### 4. Synthetic scheme for the preparation of precursor polymer and azido acid functionalized aromatic polyamides



Scheme S2. Synthetic scheme of precursor aromatic polyamide and post-polymerization modification of the precursor polymers with chiral azido acids.

### **In situ preparation of acid chloride from dicarboxylic acid (1a)<sup>3</sup>**

To dicarboxylic acid (1) (104 mg, 0.47 mmol), dry DCM (8 ml) was added under a nitrogen atmosphere, and the resulting suspension was cooled to 0°C in an ice bath. Oxalyl chloride (0.2 ml, 2.26 mmol) was added dropwise over 10 mins, followed by the addition of DMF (2 drops). The reaction mixture was stirred at room temperature for 6 hours. Excess oxalyl chloride and all the solvents were removed under reduced pressure. Assuming quantitative transformation to acid chloride, the remaining solids were directly used for the next step (polymerization).

### **General synthetic procedure for the preparation of precursor polymers (P1)<sup>3</sup>**

To pyrene-1,6-diamine (110 mg, 0.47 mmol), dry HMPA (2 ml) and dry NMP (1 ml) were added under nitrogen atmosphere, and the whole setup was placed at -78°C to chill the reaction mixture and in this process, the solution in the flask was frozen. The acid chloride (1a) (0.47 mmol), prepared from corresponding dicarboxylic acid (1), was transferred to the reaction mixture. The reaction mixture was placed in an ice-water bath and was stirred at 0 °C for 6 hours, followed by 24 hours of stirring at room temperature. After completion, DCM was removed from the reaction mixture under reduced pressure, and the residue was slowly poured into cold water. The resulting precipitates were filtered and dried. The dried solids were reprecipitated in chloroform to remove unreacted starting materials, low molecular weight species (small oligomers), etc. The remaining solids were filtered, washed with diethyl ether, and dried. This process was repeated 3 times to get the desired polymer (P1) as a brown solid.

P1 (yield: 70%)

<sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>) δ 11.80 (s, 2H, Ar-NHCO-), 8.26-8.51 (m, 8H, Ar (Pyrene)-H), 8.0 (s, Ar-H, o-OCH<sub>2</sub>-), 5.26 (d, 2H, -OCH<sub>2</sub>CCH), 3.80 (t, 1H, -OCH<sub>2</sub>CCH).

### **Post-polymerization modification (Azide-alkyne click reaction) of precursor polymers with azido acids**

To a mixture of parent polyamide (P1) (49 mg, 0.12 mmol, w.r.t to repeat unit) and Phe(L or D or D/L)-N<sub>3</sub> (67 mg, 0.35 mmol), DMSO (3 ml) was added and the reaction mixture was purged with nitrogen for 30 mins to remove dissolved oxygen from the solution. Sodium ascorbate (9 mg, 0.05 mmol) (dissolved in 30 μL of H<sub>2</sub>O) and CuSO<sub>4</sub>·5H<sub>2</sub>O (6 mg, 0.02 mmol) (dissolved in 20 μL of H<sub>2</sub>O) were added simultaneously under a nitrogen atmosphere, and the content was stirred at 50 °C for 3 days. After completion, the reaction mixture was added to diethyl ether to remove DMSO. The resulting precipitates were washed with methanol and diethyl ether, respectively. This process was repeated a few times, and the remaining solid was dried under a vacuum to get the desired product as a dark brown solid.

**P1-Phe(L)** (yield: 55%)

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  11.74 (d, 2H,  $-\text{NHCO}-$ ), 8.62 – 7.79 (m, 11H, triazole ring; Ar (Pyrene)-H; Ar (Pyridine)-H), 7.13 (d, 5H, Ar (Benzene)-H), 5.78 (s, 1H,  $-\text{CHCHH}'-$ ), 5.52 (s, 2H,  $-\text{OCH}_2\text{CCH}$ ), 3.52 (s, 2H,  $-\text{CHH}'\text{Ar}$  (Benzene)).

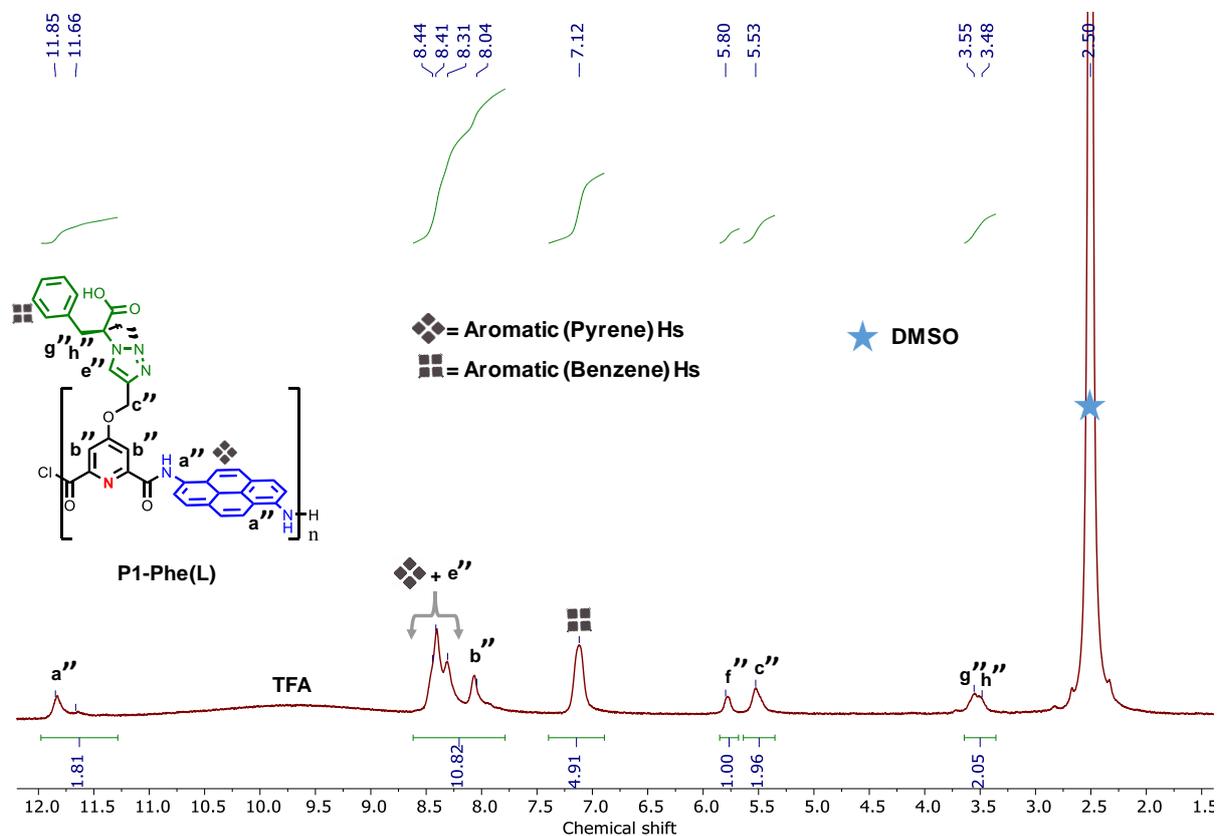


Figure S1.  $^1\text{H}$  NMR spectra of the post-polymerization modified polymer (P1-Phe(L)) [NMR spectra was recorded in  $\text{DMSO-}d_6$  and trifluoroacetic acid- $d$  (TFA- $d$ ) mixture at room temperature].

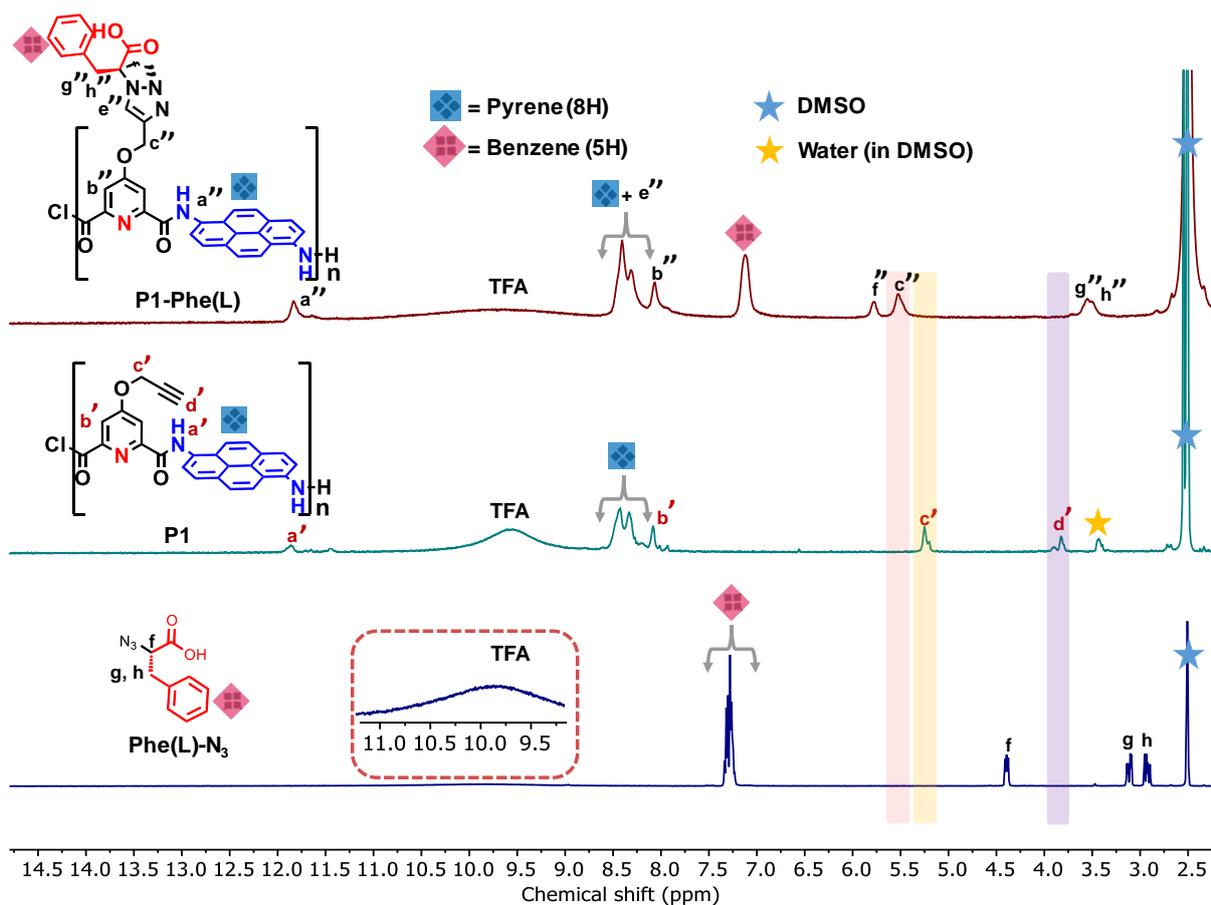


Figure S2. Stack plot  $^1\text{H}$  NMR spectra of azido acid (Phe(L)-N<sub>3</sub>) (bottom), precursor polymer (P1), and functionalized polymer (P1-Phe(L)) (top) [All the NMR spectra were recorded in a mixture of DMSO- $d_6$  and trifluoroacetic acid- $d$  (TFA- $d$ ) at room temperature. A zoomed NMR spectra (8.5 –12 ppm) of Phe(L)-N<sub>3</sub>, showcasing the peak for TFA has been depicted as the inset].

**P1-Phe(D)** (yield: 63%)

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  11.72 (d, 2H,  $-\text{NHCO}-$ ), 8.60 – 7.78 (m, 11H, triazole ring; Ar (Pyrene)-H; Ar (Pyridine)-H), 7.11 (d, 5H, Ar (Benzene)-H), 5.79 (s, 1H,  $-\text{CHCHH}'-$ ), 5.53 (s, 2H,  $-\text{OCH}_2\text{CCH}$ ), 3.50 (s, 2H,  $-\text{CHH}'\text{Ar}$  (Benzene)).

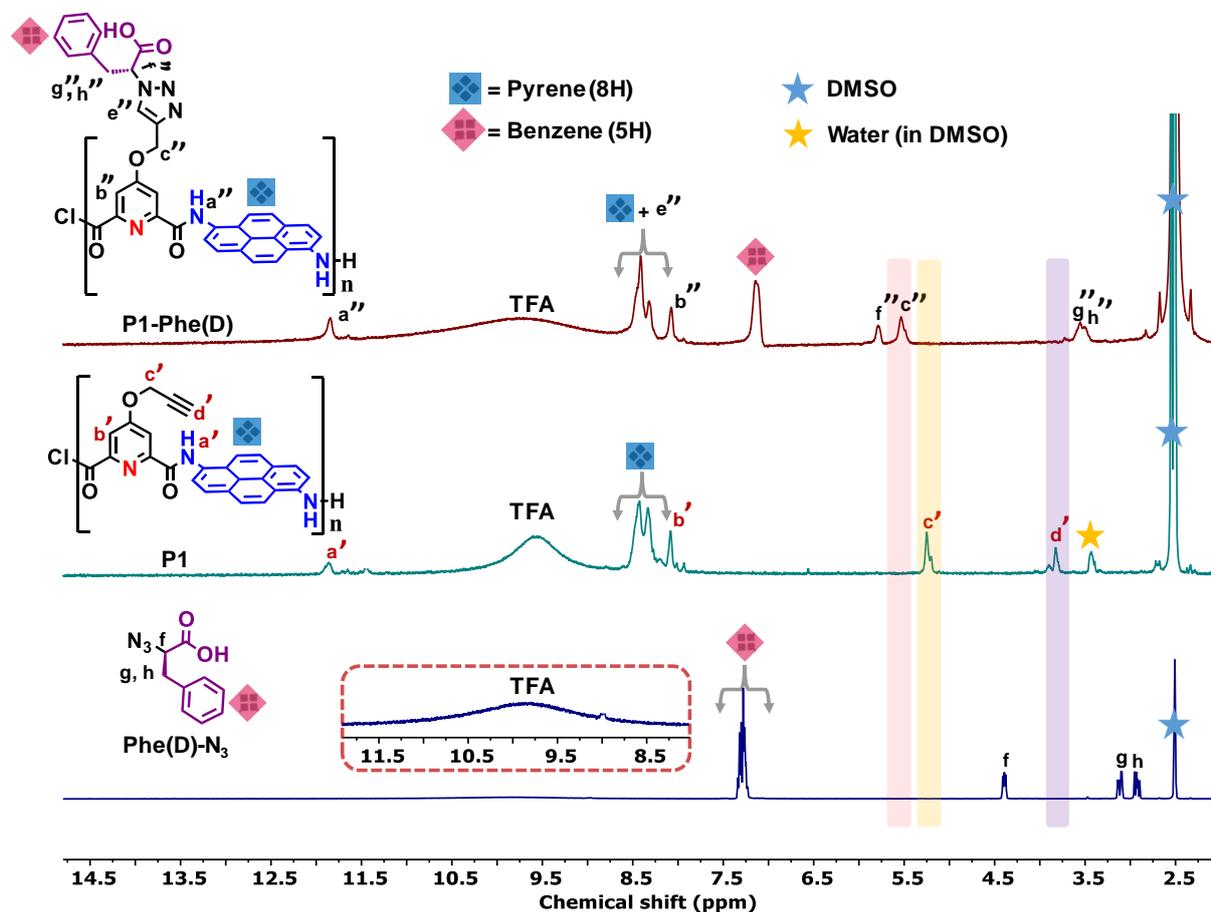


Figure S3. Stack plot  $^1\text{H}$  NMR spectra of azido acid ( $\text{Phe(D)-N}_3$ ) (bottom), precursor polymer ( $\text{P1}$ ), and functionalized polymer ( $\text{P1-Phe(D)}$ ) (top) [All the NMR spectra were recorded in a mixture of  $\text{DMSO-}d_6$  and trifluoroacetic acid- $d$  ( $\text{TFA-}d$ ) at room temperature. A zoomed NMR spectra (8.2 – 12 ppm) of  $\text{Phe(D)-N}_3$ , showcasing the peak for TFA has been depicted as the inset].

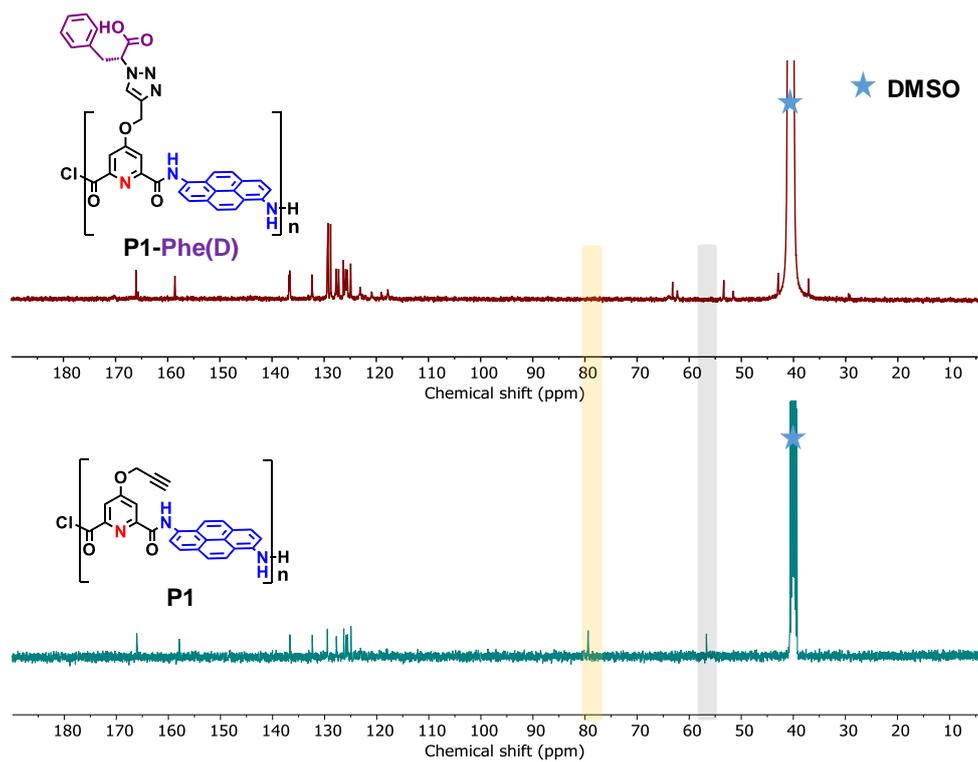


Figure S4. Stack plot <sup>13</sup>C NMR spectra of precursor polymer (P1) (bottom), and post-polymerization modified aromatic polyamide (P1-Phe(D)) (top).

### P1-Phe(D/L) (yield: 59%)

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  11.70 (d, 2H,  $-\text{NHCO}-$ ), 8.63 – 7.77 (m, 11H, triazole ring; Ar (Pyrene)-H; Ar (Pyridine)-H), 7.12 (d, 5H, Ar (Benzene)-H), 5.80 (s, 1H,  $-\text{CHCHH}'-$ ), 5.54 (s, 2H,  $-\text{OCH}_2\text{CCH}$ ), 3.49 (s, 2H,  $-\text{CHH}'\text{Ar}$  (Benzene)).

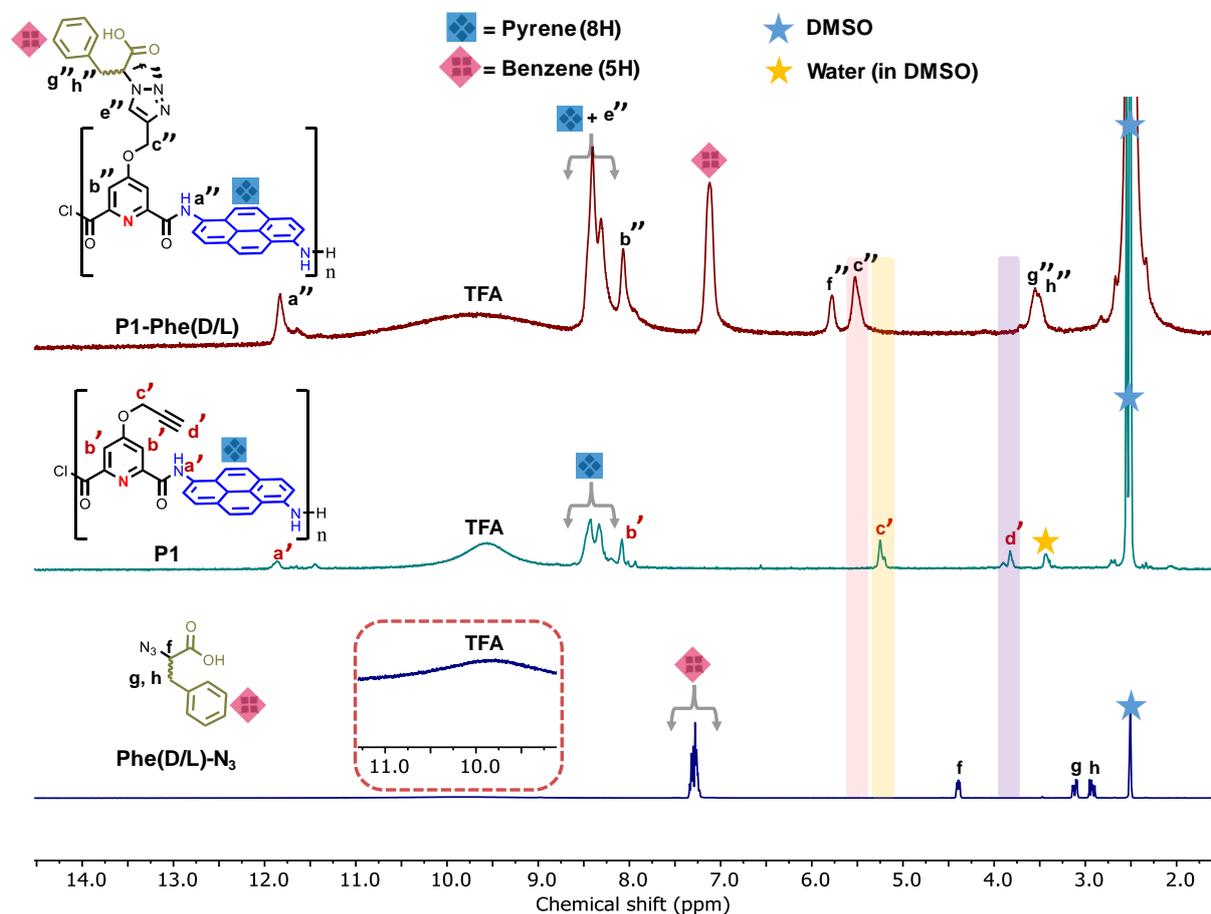


Figure S5. Stack plot  $^1\text{H}$  NMR spectra of azido acid ( $\text{Phe(D/L)-N}_3$ ) (bottom), precursor polymer ( $\text{P1}$ ), and functionalized polymer ( $\text{P1-Phe(D/L)}$ ) (top) [All the NMR spectra were recorded in a mixture of  $\text{DMSO-}d_6$  and trifluoroacetic acid- $d$  ( $\text{TFA-}d$ ) at room temperature. A zoomed NMR spectra (9 – 12 ppm) of  $\text{Phe(D/L)-N}_3$ , showcasing the peak for TFA has been depicted as the inset].

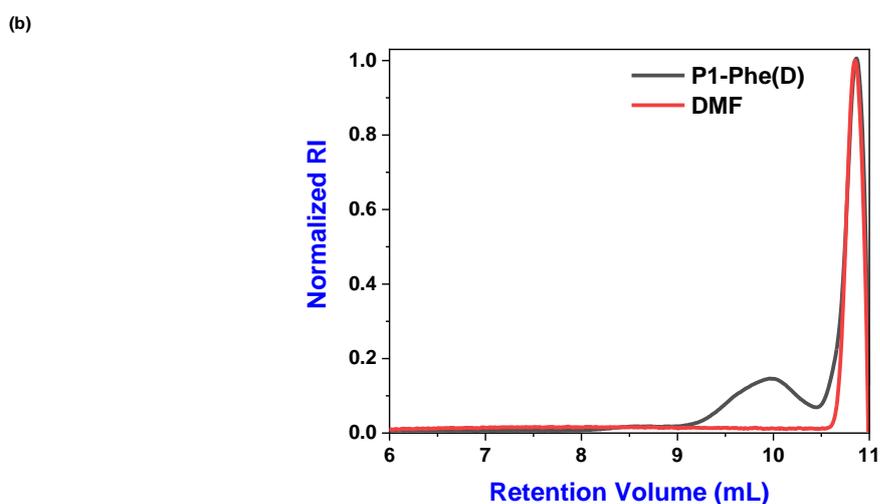
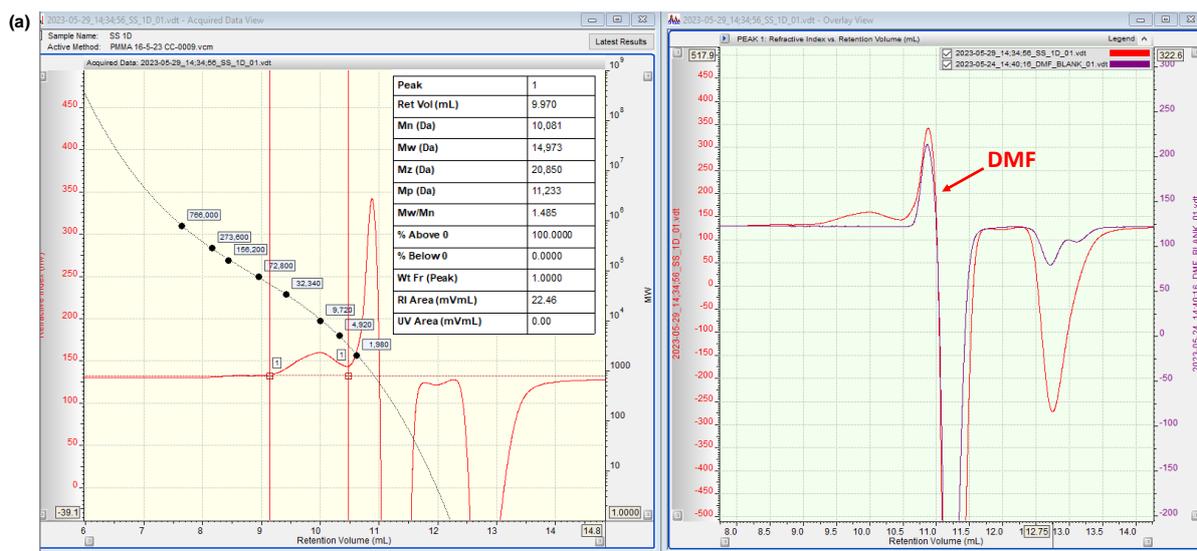
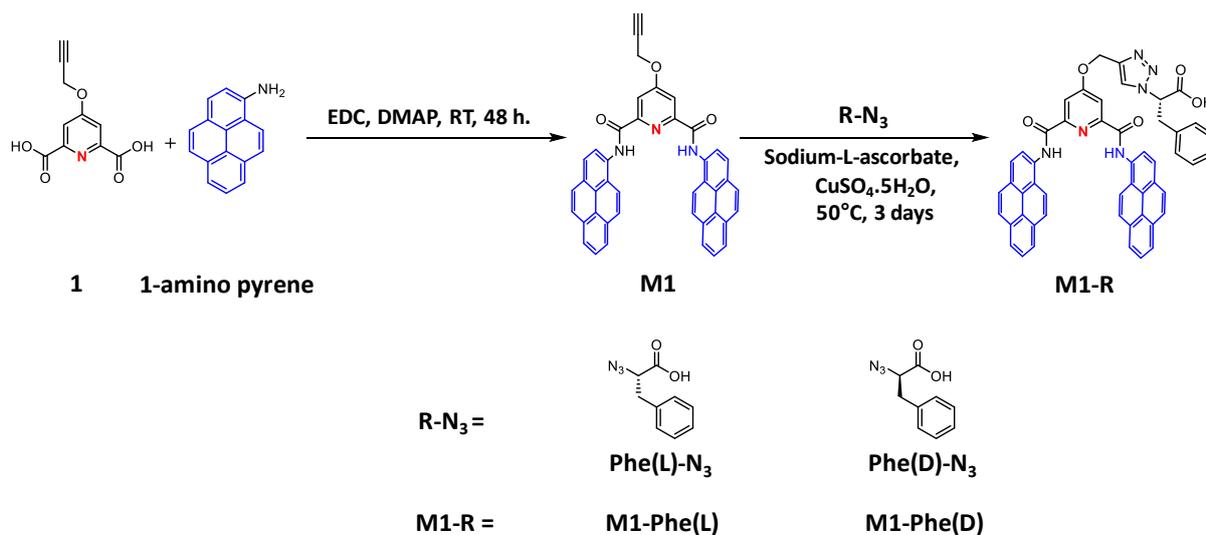


Figure S6. (a) SEC trace of P1-Phe(D) (plot using instrument software); (b) stack plot of SEC trace of P1-Phe(D) along with blank DMF (plot using Origin software).

Table S1. Table containing number average ( $M_n$ ) molecular weight, and polydispersity index ( $\mathcal{D}$ ) of the polymers.

Polymer	Mol. Wt. ( $M_n$ , gm/mole)	Dispersity ( $\mathcal{D}$ )
P1-Phe(D)	10,100	1.48
P1-Phe(L)	11,300	2.20
P1-Phe(D/L)	9,200	1.42

## 5. Synthesis of model compounds



Scheme S3. Synthetic scheme of model compounds.

### M1

To a mixture of **1** (100 mg, 0.45 mmol), 1-amino pyrene (250 mg, 1.14 mmol), and DMAP (12 mg, 0.1 mmol), dry DCM was added under a nitrogen atmosphere, and the resulting solution was cooled to 0°C in an ice bath. EDC (211 mg, 1.13 mmol) (dissolved in dry DCM) was added dropwise over 10 mins, and the reaction mixture was stirred at room temperature for 48 hours. The resulting precipitates were filtered, washed multiple times with DCM, and dried under vacuum to afford M1 as a light-yellow solid (yield: 50%).

<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 11.86 (s, 2H, -NHCO-), 8.45 – 8.06 (m, 20H, Ar (Pyrene)-H; Ar (Pyridine)-H), 5.25 (d, *J* = 2.5 Hz, 2H, -OCH<sub>2</sub>CCH), 3.85 – 3.81 (m, 1H, -OCH<sub>2</sub>CCH).

### M1-Phe(L or D)

To a mixture of M1 (50 mg, 0.08 mmol) and Phe(L or D)-N<sub>3</sub> (46 mg, 0.24 mmol), DMSO (2 ml) was added, and the reaction mixture was purged with nitrogen for 30 mins to remove dissolved oxygen from the solution. Sodium ascorbate (9 mg, 0.047 mmol) (dissolved in 30 μL H<sub>2</sub>O) and CuSO<sub>4</sub>·5H<sub>2</sub>O (6 mg, 0.024 mmol) (dissolved in 20 μL H<sub>2</sub>O) were added simultaneously under a nitrogen atmosphere, and the mixture was stirred at 50°C for 3 days. After completion, the solution was added to diethyl ether to remove DMSO. The resulting precipitates were washed with methanol and diethyl ether, respectively. This process was repeated a few times, and the remaining solids were dried under a vacuum to obtain the desired product (M1-Phe(L or D)) as a yellow solid.

**M1-Phe(L)** (yield: 51%)

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  11.86 (s, 2H, -NHCO-), 8.55 – 8.05 (m, 21H, triazole ring; Ar (Pyrene)-H; Ar (Pyridine)-H), 7.13 (s, 5H, Ar (Benzene)-H), 5.70 (s, 1H, -CHCHH'-), 5.56 (s, 2H, -OCH<sub>2</sub>CCH), 3.50 (d, 2H, -CHH'Ar (Benzene)).

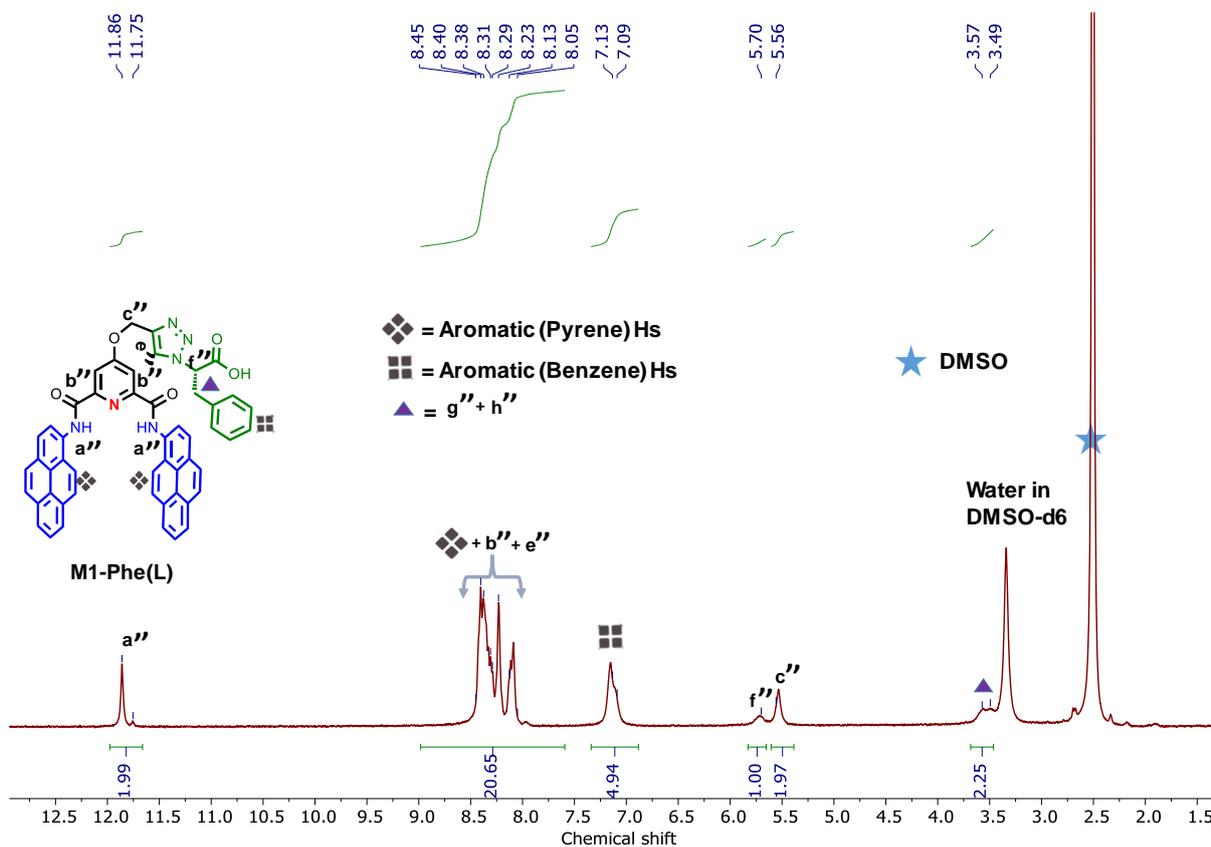


Figure S7.  $^1\text{H}$  NMR spectra of the model compound (M1-Phe(L)) [NMR spectra were recorded in  $\text{DMSO-}d_6$  at room temperature].

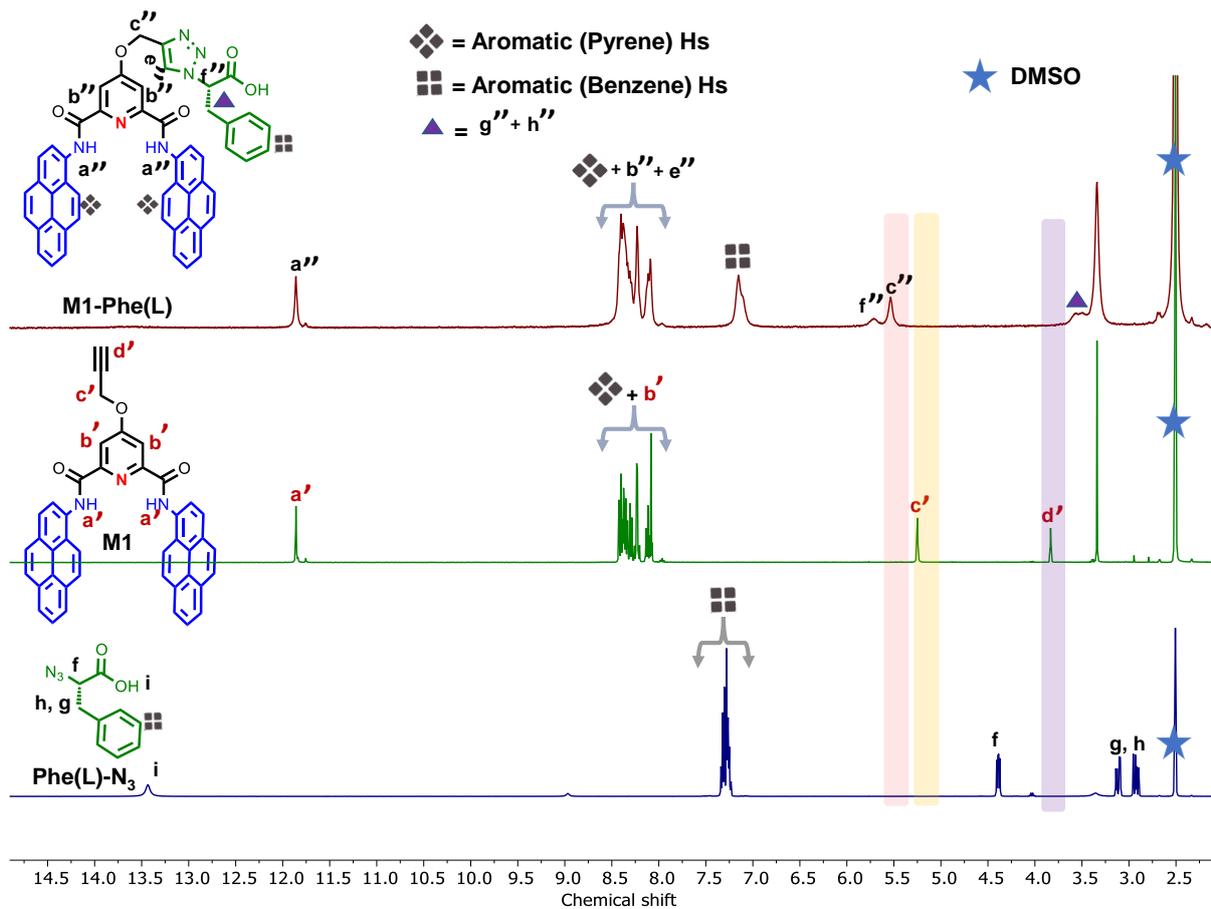


Figure S8. Stack plot  $^1\text{H}$  NMR spectra of azido acid (Phe(L)-N<sub>3</sub>) (bottom), M1, and model compound (M1-Phe(L)) (top) [NMR spectra were recorded in DMSO- $d_6$  at room temperature].

**M1-Phe(D)** (yield: 53%)

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  11.86 (s, 2H, -NHCO-), 8.55 – 7.98 (m, 21H, triazole ring; Ar (Pyrene)-H); Ar (Pyridine)-H), 7.15 (s, 5H, Ar (Benzene)-H), 5.72 (s, 1H, -CHCHH'-), 5.54 (s, 2H, -OCH<sub>2</sub>CCH), 3.52 (d, 2H, -CHH'Ar (Benzene)).

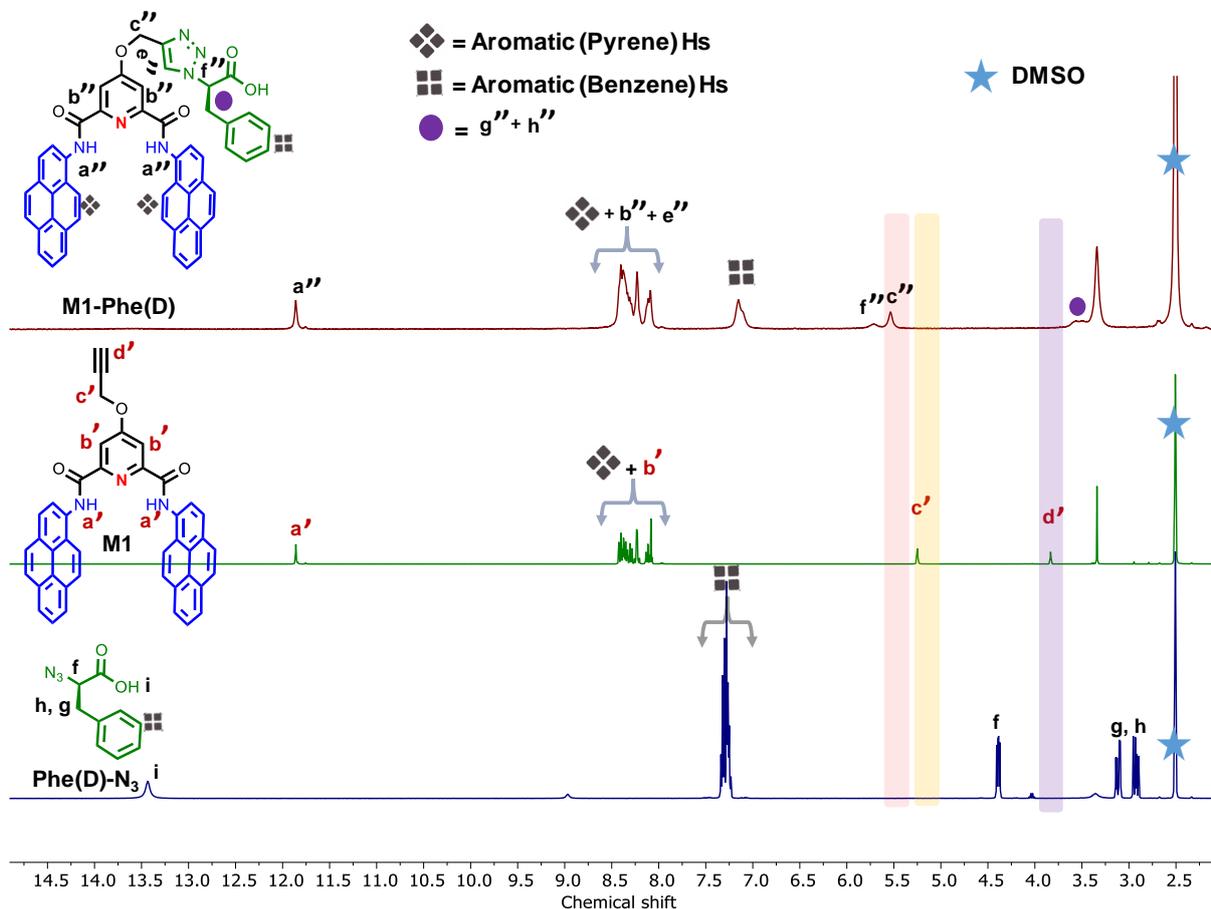


Figure S9. Stack plot  $^1\text{H}$  NMR spectra of azido acid ( $\text{Phe(D)-N}_3$ ) (bottom), M1, and model compound (M1-Phe(D)) (top) [NMR spectra were recorded in  $\text{DMSO-}d_6$  at room temperature].

## 6. Concentration-dependent UV-Vis and CD studies

Before conducting the folding analysis, concentration-dependent UV-Vis and circular dichroism (CD) experiments were performed to ensure the single-chain behavior of the polymer chains at the working concentration (50  $\mu\text{M}$ ) in a solvent mixture of DMSO and triethylamine (TEA) (99.5 : 0.5)

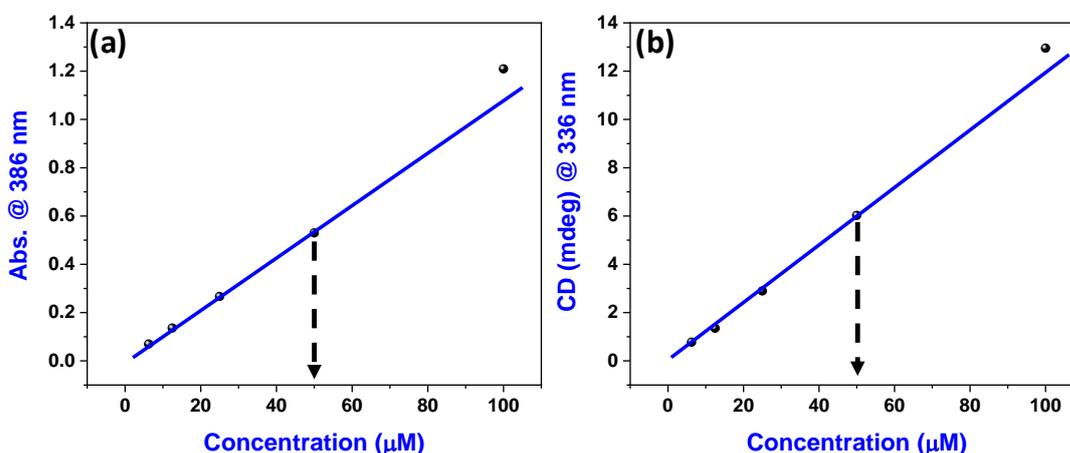


Figure S10. Variation of UV-Vis absorption (@386 nm) (a) and CD intensity (@336 nm) (b) of P1-Phe(D) with concentration. The linear variation at the working concentration (50  $\mu\text{M}$ , marked with an arrow) was observed.

## 7. Helicity induction and single handedness

The samples for the photophysical and chiroptical studies were prepared in a solvent mixture of DMSO and triethylamine (TEA). The solid samples were added to the solvent mixture (DMSO + triethylamine (TEA) (99.5 : 0.5) and allowed to solubilize at room temperature. These solutions were then heated to 50  $^{\circ}\text{C}$  followed by slow cooling (30 min), and the process was repeated for another cycle before being used for further studies.

Table S2. Table containing estimated size and  $k_{\text{cps}}$  of the polymers using dynamic light scattering.

Polymer	Size (nm)	$k_{\text{cps}}$
P1-Phe(D)	$2.5 \pm 0.5$	3.8
P1-Phe(L)	$2.9 \pm 0.4$	4.9
P1-Phe(D/L)	$2.2 \pm 0.3$	4.8

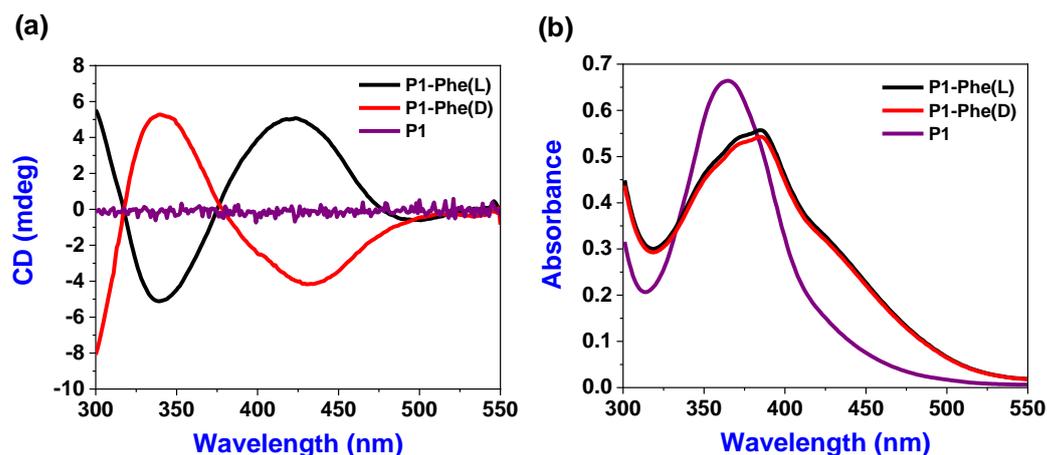


Figure S11. (a) CD spectra of P1-Phe(L), P1-Phe(D), and precursor polyamide (P1); (b) UV-Vis spectra of P1-Phe(L), P1-phe(D), precursor polyamide (P1) [All the solutions were prepared in DMSO + triethylamine (TEA) (99.5 : 0.5) at 50  $\mu$ M].

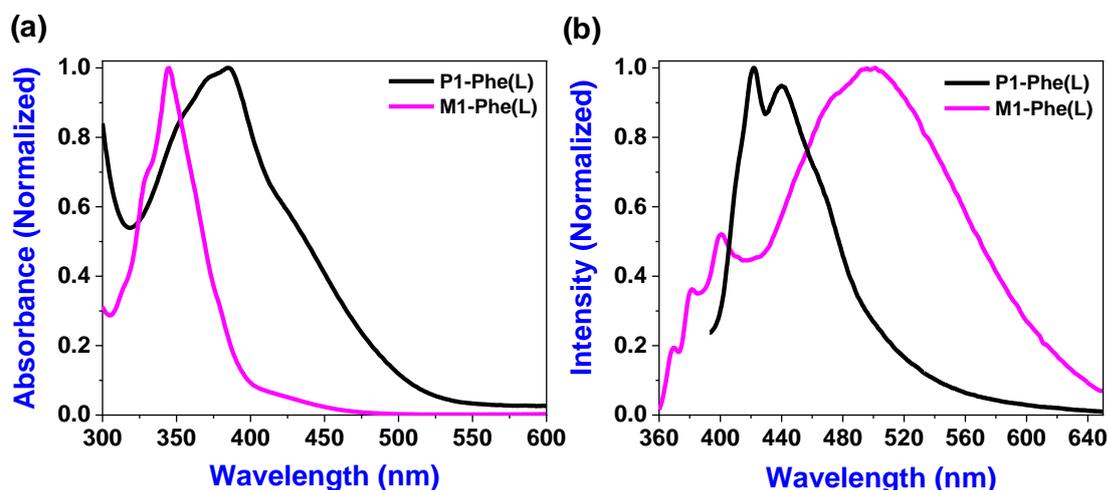


Figure S12. (a) Normalized UV-Vis spectra of P1-Phe(L) and the model compound (M1-Phe(L)); (b) Normalized fluorescence spectra of P1-Phe(L) and the model compound (M1-Phe(L)) [All the solutions were prepared in DMSO + triethylamine (TEA) (99.5 : 0.5) at 50  $\mu$ M].

## 8. TEM Imaging

The samples of the TEM imaging were prepared by dropcasting the dilute (50  $\mu\text{M}$ ) polymer solution (in DMSO + triethylamine (TEA) (99.5 : 0.05) mixture) on the carbon-coated Cu grids and was followed by drying under vacuum for more than 12 hours.

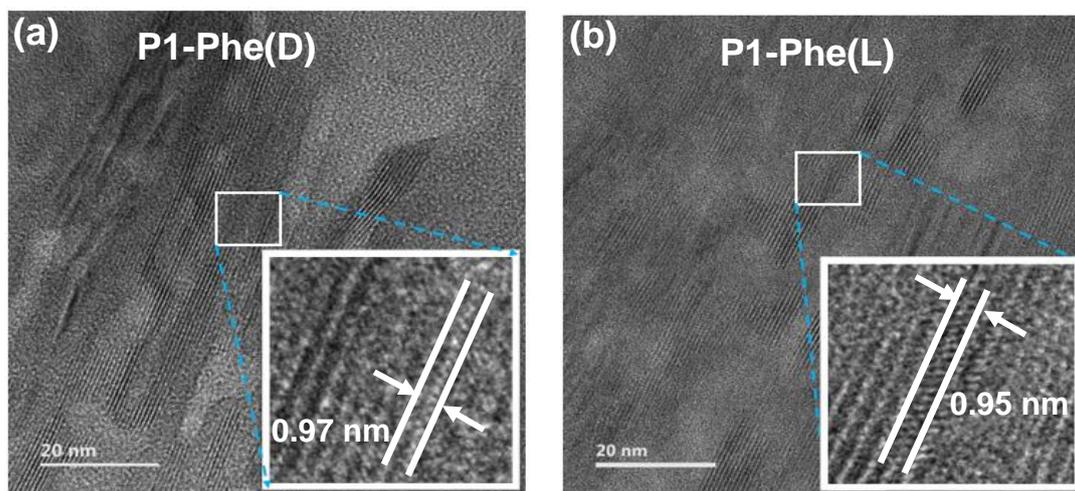
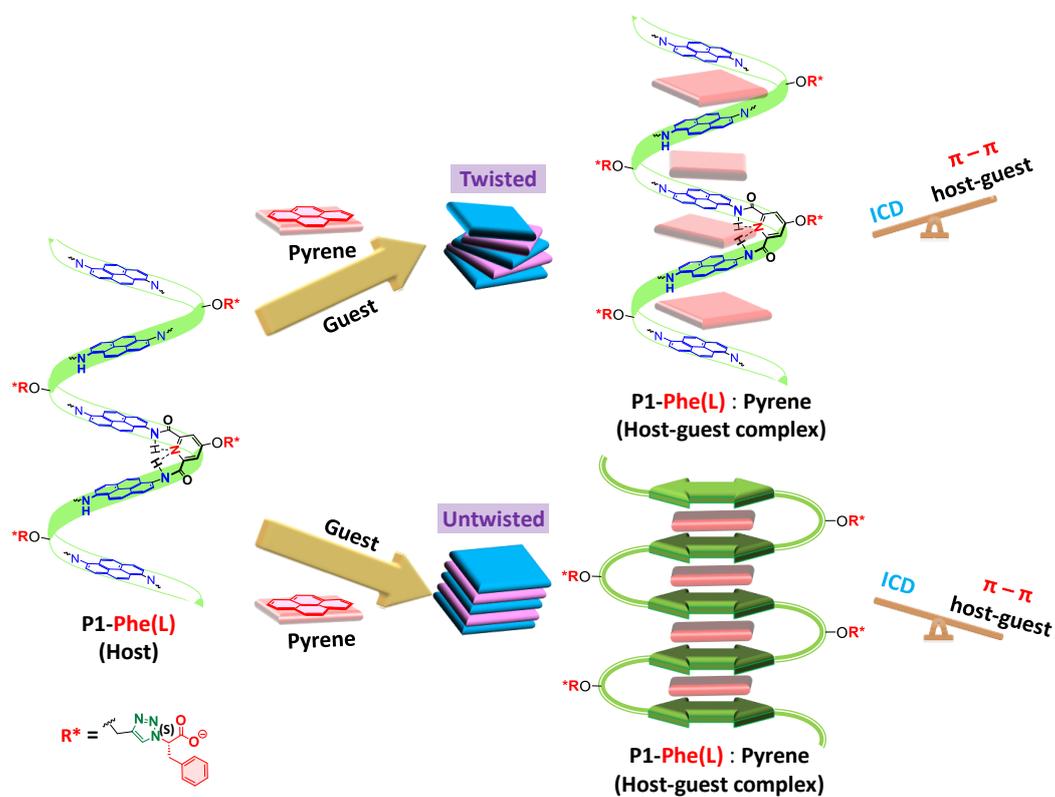


Figure S13. HRTEM images of P1-Phe(D) (a) and P1-Phe(L) (b) respectively; inset showing the zoomed images of the helix structures, mentioning their chain-to-chain spacing.

## 9. Effect of the guest molecule on the helical folding



Scheme S4. A schematic representation depicting the possible host-guest complexation of pyrene (guest) within the twisted aromatic polyamide scaffold (host).

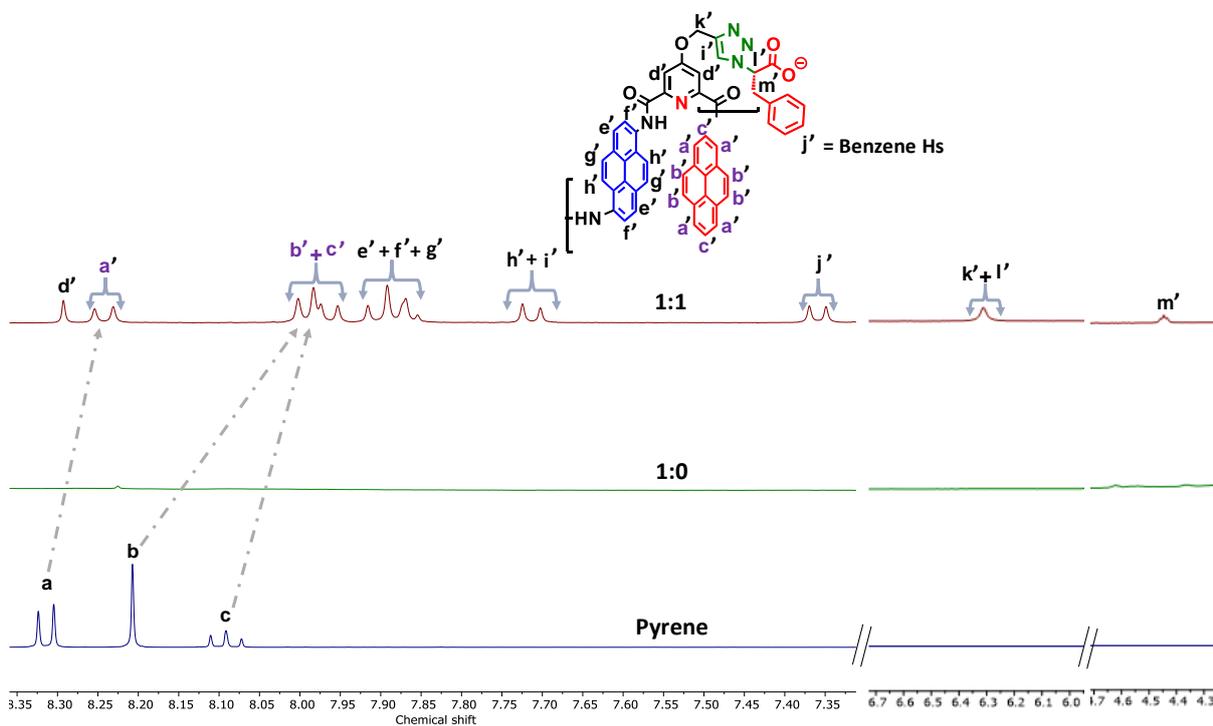
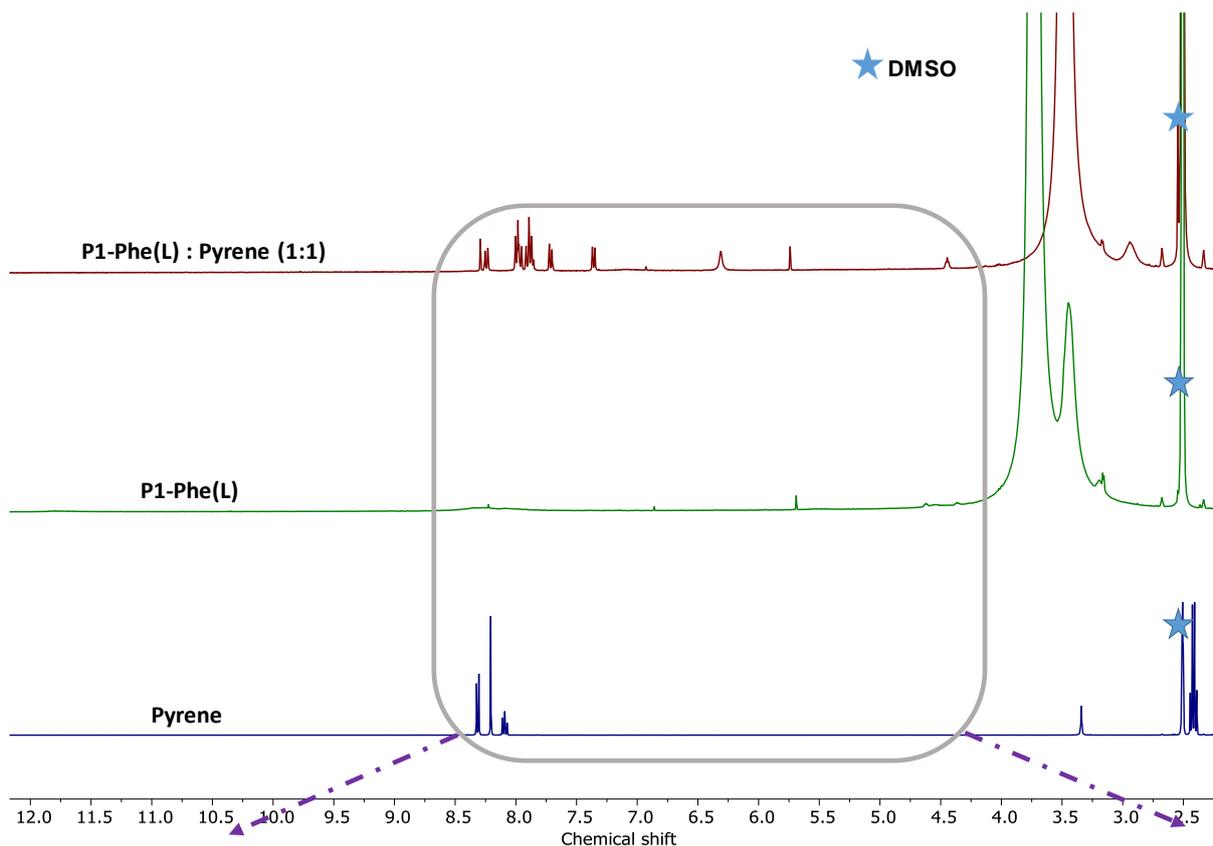


Figure S14. Stack plot of the  $^1\text{H}$  NMR (400 MHz) spectra of pyrene (bottom), P1-Phe(L) polymer (middle), and 1:1 host-guest complex of P1-Phe(L):pyrene (top) [  $^1\text{H}$  NMR spectra were recorded in a mixture of DMSO- $d_6$  and triethylamine (TEA) (99.5:0.5) at 1 mM].

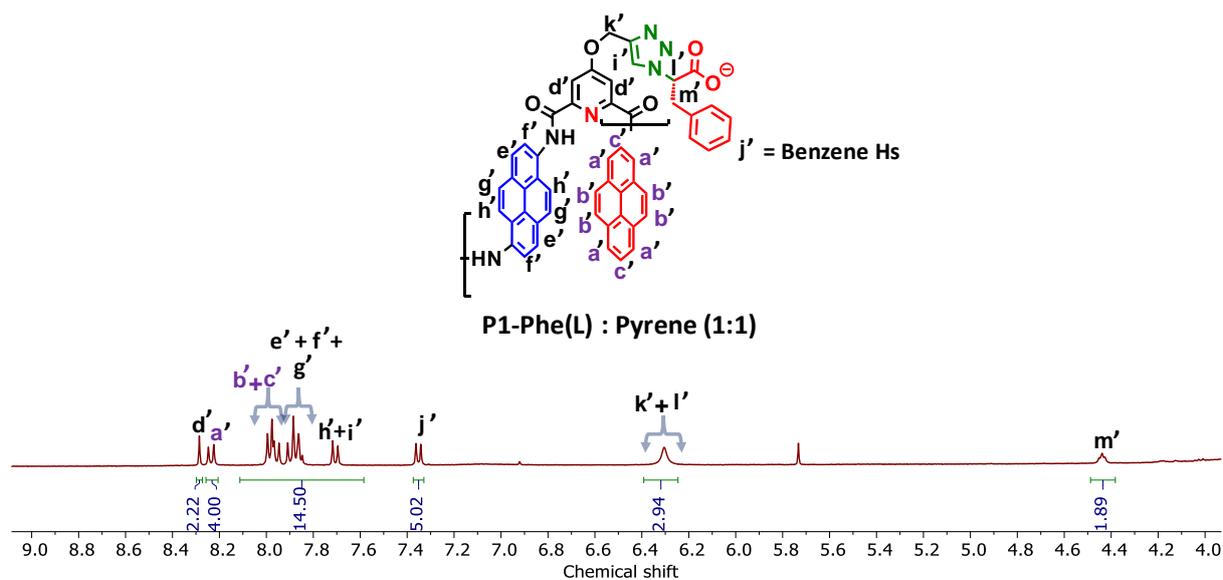


Figure S15.  $^1\text{H}$  NMR (400 MHz) assignments of 1:1 host-guest complex of P1-Phe(L):pyrene [  $^1\text{H}$  NMR spectra were recorded in a mixture of DMSO- $d_6$  and triethylamine (TEA) (99.5:0.5) at 1 mM].

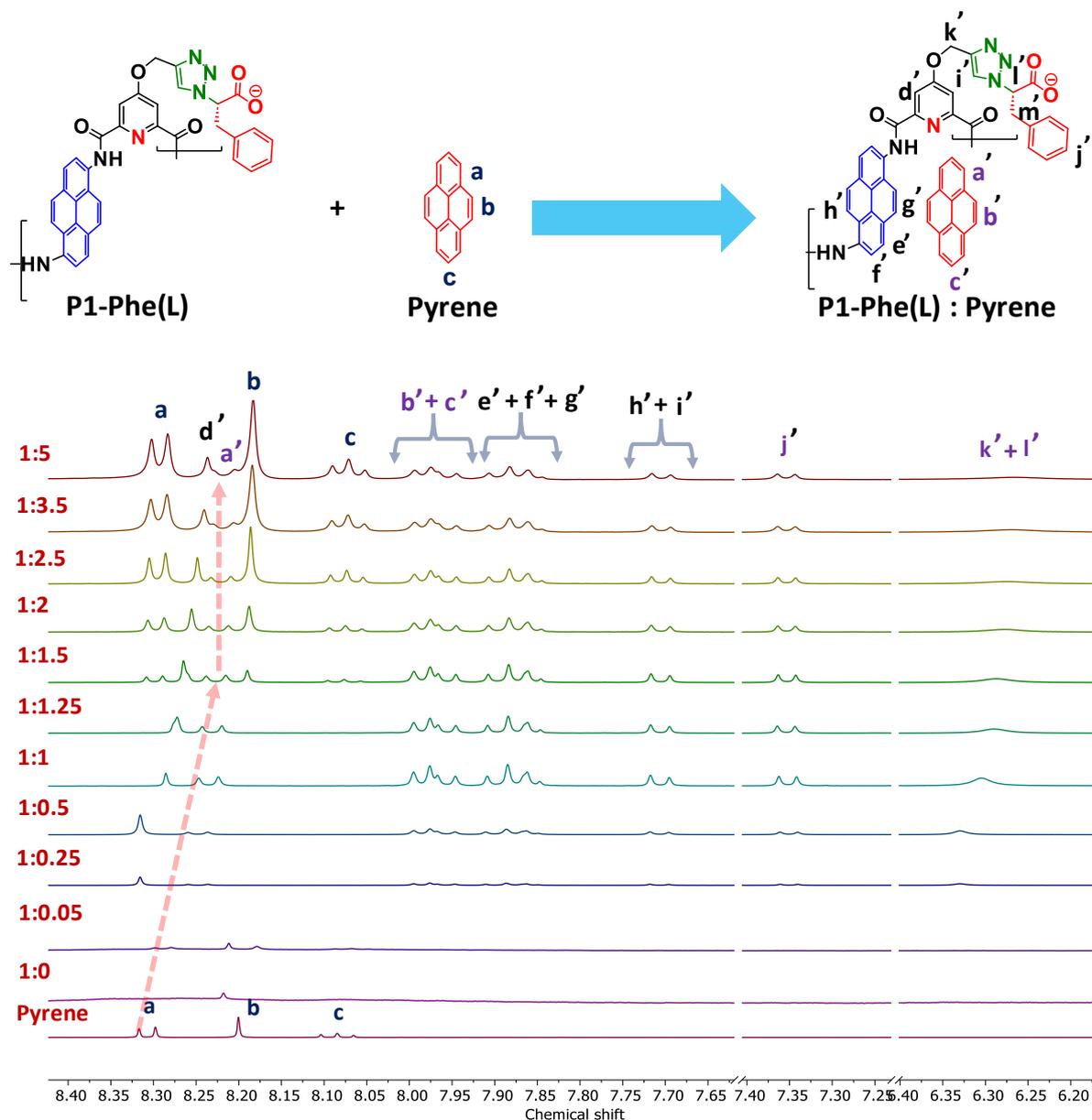


Figure S16. A stacked plot of the  $^1\text{H}$  NMR spectra (400 MHz) of pyrene, P1-Phe(L) (1:0), and P1-Phe(L):pyrene (host-guest) complex, demonstrating the spectral changes upon addition of increasing amount of pyrene to the polymer scaffold; increasing the pyrene concentration beyond 1 equivalent results in the appearance of uncomplexed pyrene peaks around their original positions, indicated by the asterisks (\*) [ $^1\text{H}$  NMR spectra were recorded in a mixture of DMSO- $d_6$  and triethylamine (TEA) (99.5 : 0.5)) at 1 mM].

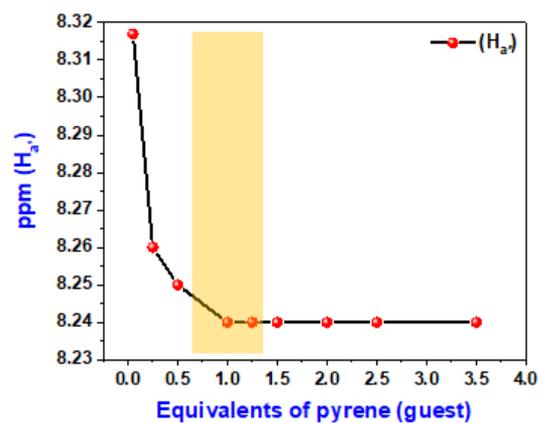


Figure S17. A titration plot of the chemical shift values of pyrene proton in the host-guest complex (P1-Phe(L):pyrene; at ~8.32 ppm, marked a') as a function of added equivalents of pyrene.

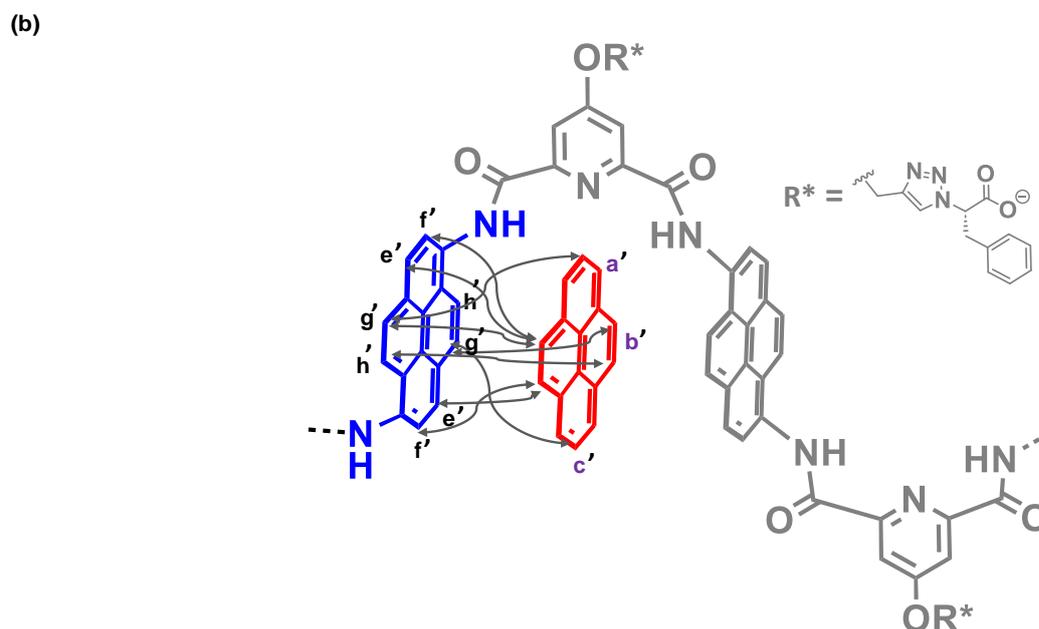
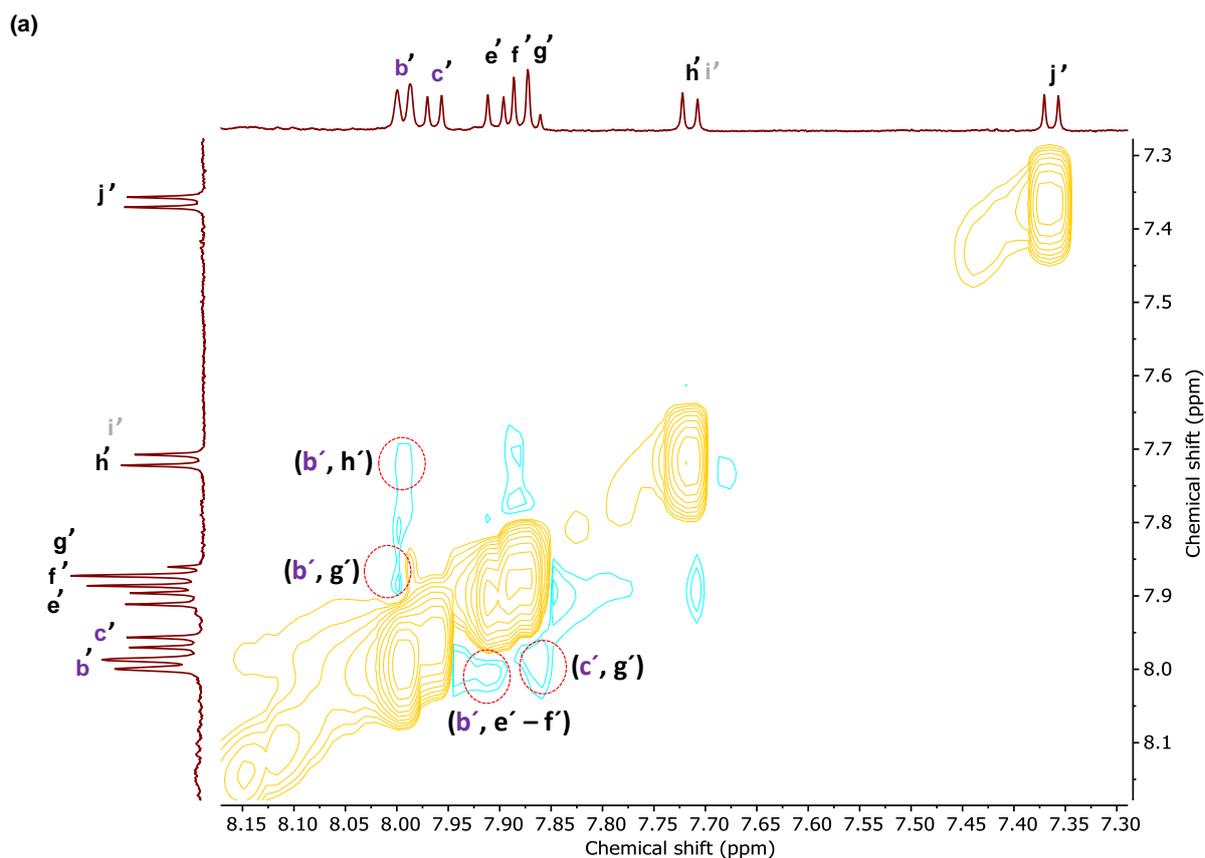
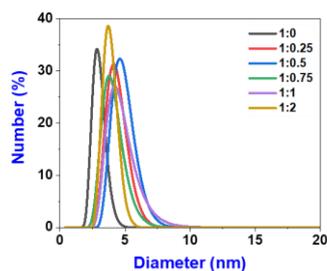


Figure S18. (a) Partial NOESY spectra (600 MHz) of P1-Phe(L):pyrene (1:1, host-guest) complex at room temperature reveal the NOEs between the complexed pyrene protons ( $b'$ ,  $c'$ ) and the protons of the pyrene diamide of the host polymer ( $e'$ ,  $f'$ ,  $g'$ ,  $h'$ ) [ $^1\text{H}$  NMR spectra was recorded in a mixture of DMSO- $d_6$  and triethylamine (TEA) (99.5 : 0.5) at 1 mM]; (b) illustration of the 1:1 (host-guest) complex between P1-Phe(L) and pyrene, with the major NOEs between the protons of the complexed pyrene and pyrene diamide of the polymer (double headed arrows indicate the interactions).



P1-Phe(L):pyrene	Size (nm)	kcps
1:0	2.9 ± 0.4	4.9
1:0.25	4.2 ± 0.69	4.8
1:0.5	4.7 ± 0.75	4.0
1:0.75	3.9 ± 0.7	3.5
1:1	4.4 ± 0.9	3.3
1:2	3.7 ± 0.45	6.8

Figure S19: DLS studies of host polymer and host-guest (P1-Phe(L):pyrene) complex. A small size along with low kcps values associated with DLS data is indicative of single chain behavior of these aromatic polyamides at ~50  $\mu$ M concentration.

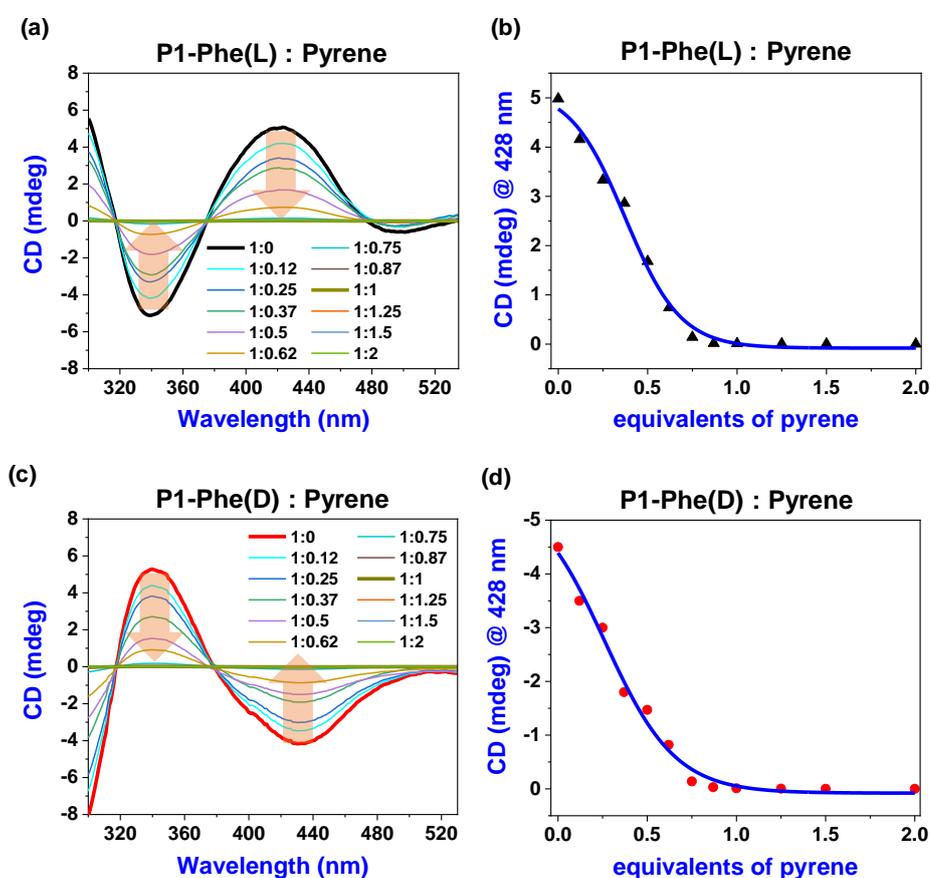


Figure S20. (a) CD spectra of P1-Phe(L):pyrene (host-guest) complex, demonstrating the spectral changes upon addition of increasing amount of pyrene to the polymer scaffold; (b) a titration plot of CD intensity (mdeg) at 428 nm of the P1-Phe(L):pyrene (host-guest) complex as a function of added equivalents of pyrene (the solid line represents the calculated fitting curve for the experimental data point series); (c) CD spectra of P1-Phe(D):pyrene (host-guest) complex, demonstrating the spectral changes upon addition of increasing amount of pyrene to P1-Phe(D); (d) a titration plot of CD intensity (mdeg) at 428 nm of the P1-Phe(D):pyrene (host-guest) complex as a function of added equivalents of pyrene (the solid line represents the calculated fitting curve for the experimental data point series) [All the solutions were prepared in DMSO + triethylamine (TEA) (99.5 : 0.5) at 50  $\mu$ M].

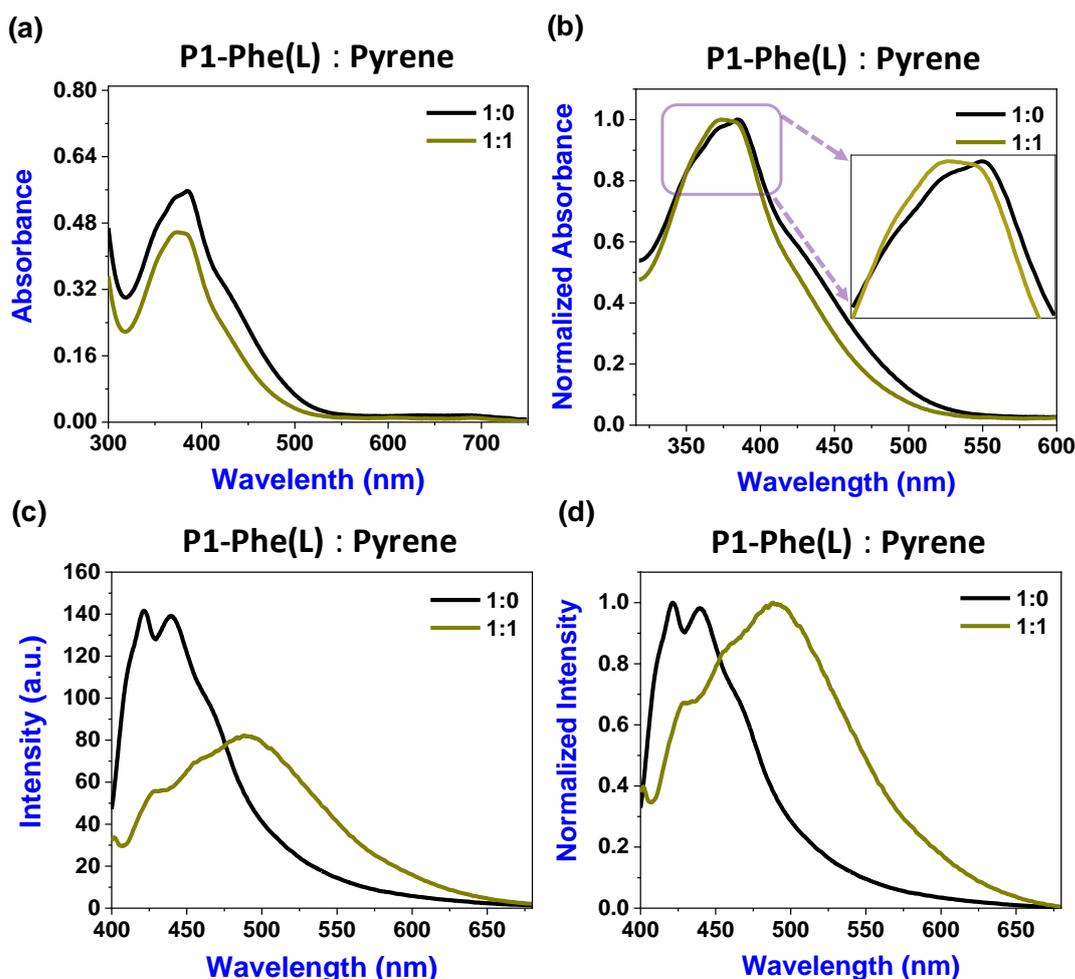


Figure S21. (a) Absorption spectra of P1-Phe(L) (1:0) and P1-Phe(L):pyrene (host-guest, 1:1) complex; (b) normalized absorption spectra of P1-Phe(L) (1:0) and P1-Phe(L):pyrene (host-guest, 1:1) complex [inset showing the zoomed absorption spectra of the polymer(1:0) and the host-guest complex (1:1)]; (c) absolute emission spectra of P1-Phe(L) (1:0) and P1-Phe(L):pyrene (host-guest, 1:1) complex; (d) normalized emission spectra of P1-Phe(L) (1:0) and P1-Phe(L):pyrene (host-guest, 1:1) complex [All the solutions were prepared in DMSO + triethylamine (TEA) (99.5 : 0.5) at 50  $\mu$ M].

## 10. Twisting – Untwisting - Retwisting of aromatic polyamides

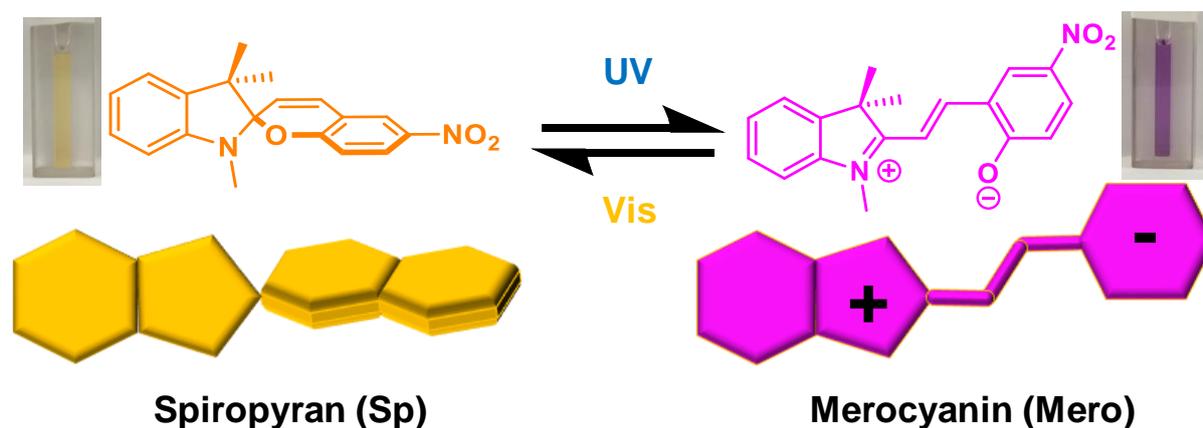


Figure S22. Reversible conversion of spiropyran (Sp) to merocyanine (Mero) upon exposure to UV and Visible light.

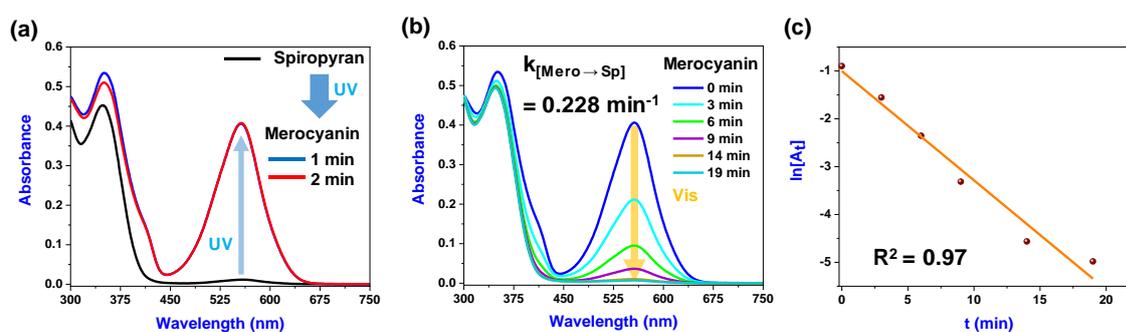


Figure S23. (a) UV-Vis spectra of spiropyran upon irradiation to UV-light (365 nm); (b) UV-Vis spectra of merocyanine at different time intervals upon exposure to visible light; (c) plot of the  $\ln[A_t]$  vs the time of the visible light irradiation (min),  $[A_t]$  represents the concentration of merocyanin at a certain time, which was determined by taking the UV-Vis absorption maxima value of the 557 nm peak of merocyanin alone [Solvent = DMSO, concentration = 50  $\mu\text{M}$ ].

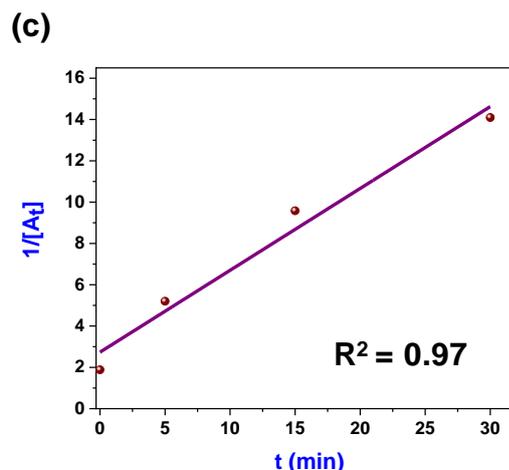
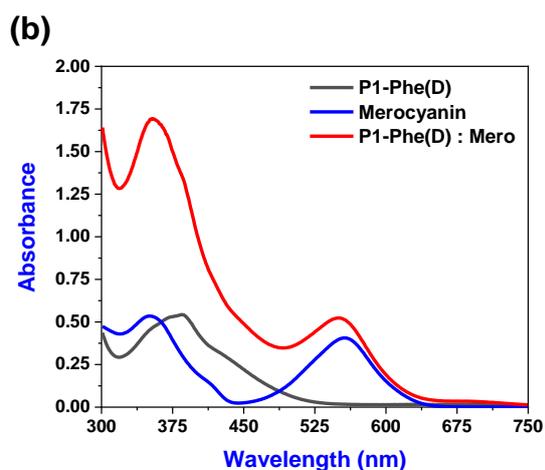
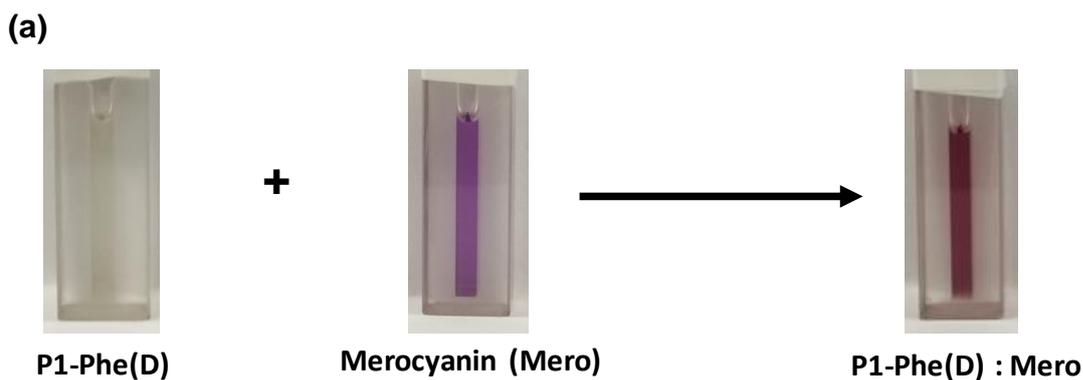


Figure S24. (a) Pictorial representation of P1-Phe(D) (host, light yellow), merocyanine (guest, pink) and the host-guest complex (P1-Phe(D):Mero, brown); (b) UV-Vis spectra of P1-Phe(D), merocyanine, and P1-Phe(D):Mero (1:1) host-guest complex; (c) plot of the  $1/[A_t]$  vs the time of the visible light irradiation (min),  $[A_t]$  represents the concentration of merocyanine at a certain time, which was determined by taking the UV-Vis absorption maxima value of the 550 nm peak of P1-Phe(D):Mero (1:1) complex.

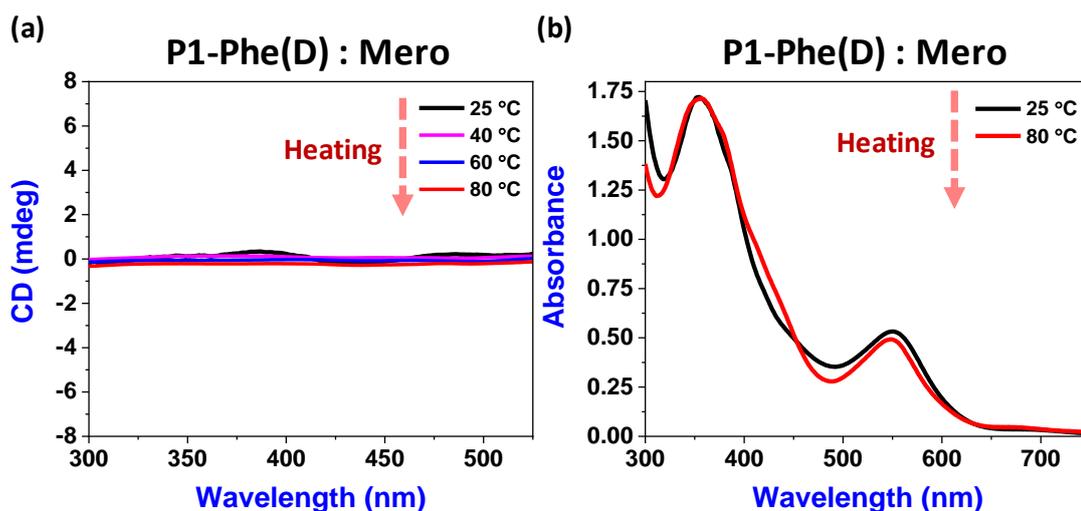


Figure S25. Thermal stability of the host polymer and their 1:1 host-guest complex (50  $\mu$ M); (a) thermal stability probed by VT-CD instrument; (b) thermal stability probed by VT-UV instrument.

## 11. References

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2. C. M. Lin, M. Arancillo, J. Whisenant and K. Burgess, *Angew. Chem. Int. Ed.*, 2020, **59**, 9398–9402.
3. S. Samanta, D. Mallick and R. K. Roy, *Polym. Chem.*, 2022, **13**, 3284–3293.