Supporting Information:

A systematic understanding of gelation self-assembly: Solvophobically assisted supramolecular gelation via conformational reorientation across amide functionality on a hydrophobically modulated dipeptide based ambidextrous gelator, N-\(n\)-acyl-(L)Val-X(Obn), (X = 1,ω-amino acid)

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Scheme:

$$\text{R} \text{O} \text{N} \text{H} \text{O} \text{R} + \text{N} \text{O} \text{O} \text{H} \text{C} \text{C} \text{R} \text{O} \text{N} \text{O} \text{O} \text{H} \text{D} \text{C} \text{C} \text{R} \text{O} \text{N} \text{O} \text{O} \text{H}$$

$$(1)$$

$$\text{EtOAc; 6 hrs} \quad >98\%$$

$$\text{Valine; NEt}_3; \quad 65\% \text{aq EtOH} \quad \text{reflux 60-80} \ C; 20\text{hrs} \quad >96\%$$

$$\text{R} \quad \text{(1eqv)} \quad \text{EtOAc; 6 hrs} \quad >98\%$$

$$\text{Valine; NEt}_3; \quad 65\% \text{aq EtOH} \quad \text{reflux 60-80} \ C; 20\text{hrs} \quad >96\%$$

$$\text{R} = \text{C}_9\text{H}_{19} \quad 2(\text{a})$$
$$\text{R} = \text{C}_{11}\text{H}_{23} \quad 2(\text{b})$$
$$\text{R} = \text{C}_{13}\text{H}_{27} \quad 2(\text{c})$$
$$\text{R} = \text{C}_{15}\text{H}_{31} \quad 2(\text{d})$$
$$\text{R} = \text{C}_{17}\text{H}_{35} \quad 2(\text{e})$$
$$\text{R} = \text{C}_7\text{H}_{15} \quad 2(\text{f})$$

$$\text{HOBT, DCC, DMAP, K}_2\text{CO}_3; \text{THF/CHCl}_3 (1:1), \text{overnight}$$

$$(\text{1eqv})$$

$$\text{H}_2\text{N}-\text{R} \text{OH} + \text{Ph-SO}_3^- \quad \text{PTSA; Reflux, 24hrs} \quad >96\%$$

$$\text{H}_2\text{N}-\text{R} \text{OCH}_2\text{Ph} \quad n = 2, 3, 5, 10$$

4(a) R = C9H19 ; n=2
4(b) R = C11H23 ; n=2
4(c) R = C13H27 ; n=2
4(d) R = C15H31 ; n=2
4(e) R = C17H35 ; n=2
4(f) R = C13H27 ; n=3
4(g) R = C13H27 ; n=5
4(h) R = C7H15 ; n=10
Synthetic procedure:

Synthesis of NHS ester of Fatty acids (1): To a solution of Fatty acid (1eqv) in 10 ml Ethyl acetate, N-hydroxysuccinimide (1eqv) was added followed by the addition of DCC (Dicyclohexylcarbodiimide) (1eqv) in 5 ml Ethyl acetate and the reaction stirred for 6-7 hrs. Completion of the reaction was confirmed by TLC. The reaction mixture was then filtered and the filtrate was concentrated under reduced pressure. The obtained residue was recrystallized from isopropanol and used for the next step without any further purification (~98% yield).

Synthesis of N-Acyl-valine (2): To a solution of NHS ester of fatty acids (8.4 mmole, 1 eqv), (L)-Valine (1.2 eqv) in 40 ml of 65%aq Ethanol; Triethylamine (2.5 eqv) was added and put to reflux (60-80°C) for 20 hrs. Then the reaction mixture was concentrated under reduced pressure and taken up in 25ml Ethyl acetate and washed sequentially with ice cold 1 (N) HCl (30 ml × 2), saturated brine solution (25 ml × 2). Organic layer was collected and dried over anhyd. Na₂SO₄ and evaporated under reduced pressure to give the desired product (~8.06mmol; ~96% yield; m.p= ~95-98°C). The compounds were used for the next step without any further purification.

2a: ¹HNMR (300 MHz,CDCl₃):-δ::0.83-0.89 (t,CH₃(CH₂)₈CONH-,3H), 0.89-0.98 (q, (CH₃)₂CH-CH (COOH)NH-, 7H), 1.1-1.3 (bs, CH₃(CH₂)₈CH₂CH₂CONH-, 12H), 1.56-1.7 (t, CH₃(CH₂)₁₂CH₂CH₂CONH-, 2H), 2.2-2.35 (t, CH₃(CH₂)₆CH₂CH₂CONH-, 2H), 4.55-4.64 (q, -NHCH (COOH)CH(CH₃)₂, 1H), 5.89-5.99 (d, -NH₂, 1H).

2b: ¹HNMR (300 MHz,CDCl₃):-δ::0.88-0.92 (t,CH₃(CH₂)₁₀CONH-,3H), 0.89-1.0 (q, (CH₃)₂CH-CH (COOH)NH-, 7H), 1.2-1.44 (bs, CH₃(CH₂)₆CH₂CH₂CONH-, 16H), 1.56-1.7 (t, CH₃(CH₂)₁₂CH₂CH₂CONH-, 2H), 2.21-2.35 (t, CH₃(CH₂)₆CH₂CH₂CONH-, 2H), 4.55-4.61 (q, -NHCH (COOH)CH(CH₃)₂, 1H), 5.89-5.99 (d, -NH₂, 1H).

2c: ¹HNMR (300 MHz,CDCl₃):-δ::0.86-0.91 (t,CH₃(CH₂)₁₁CONH-,3H), 0.89-1.0 (q, (CH₃)₂CH-CH (COOH)NH-, 7H), 1.2-1.45 (bs, CH₃(CH₂)₁₀CH₂CH₂CONH-, 20H), 1.56-1.7 (t, CH₃(CH₂)₁₂CH₂CH₂CONH-, 2H), 2.23-2.35 (t, CH₃(CH₂)₁₁CH₂CH₂CONH-, 2H), 4.55-4.621 (q, -NHCH (COOH)CH(CH₃)₂, 1H), 5.89-5.99 (d, -NH₂, 1H).

2d: ¹HNMR (300 MHz,CDCl₃):-δ::0.89-0.89 (t,CH₃(CH₂)₁₂CONH-,3H), 0.89-1.0 (q, (CH₃)₂CH-CH (COOH)NH-, 7H), 1.25-1.44 (bs, CH₃(CH₂)₁₂CH₂CH₂CONH-, 24H), 1.56-1.7 (t, CH₃(CH₂)₁₂CH₂CH₂CONH-, 2H), 2.22-2.35 (t, CH₃(CH₂)₁₂CH₂CH₂CONH-2, H), 4.55-4.61 (q, -NHCH (COOH)CH(CH₃)₂, 1H), 5.89-5.99 (d, -NH₂, 1H).

2e: ¹HNMR (300 MHz,CDCl₃):-δ::0.91-1.10 (t,CH₃(CH₂)₁₆CONH-,3H), 0.89-1.0 (q, (CH₃)₂CH-CH (COOH)NH-, 7H), 1.2-1.46 (bs, CH₃(CH₂)₁₄CH₂CH₂CONH-, 28H), 1.56-1.7 (t, CH₃(CH₂)₁₂CH₂CH₂CONH-, 2H), 2.23-2.35 (t, CH₃(CH₂)₁₄CH₂CH₂CONH-, 2H), 4.55-4.611 (q, -NHCH (COOH)CH(CH₃)₂, 1H), 5.89-5.99 (d, -NH₂, 1H).

Synthesis of p-toluenesulfonatesalcohol 1-ω-Aminoacid benzylester (3): In 100ml of benzene a mixture of 1,ω-Aminoacid (1 eqv), p-toluenesulfonic acid (PTSA).H₂O (1.1 eqv) and benzyl alcohol (2 eqv) was taken. After refluxing in Dean-Stark apparatus for 24hrs, 40 ml of diethyl ether was added to it with immediate precipitation of PTSA-salt of 1-ω-Aminoacid benzyl ester. The salt was then filtered, washed
with 10ml of dry diethyl ether and dried under reduced pressure. This compound was used for the next step without any further purification. (yield~96%)

**Synthesis of (4):** To a solution of N-Acyl-(L)-valine (1 eqv) in 5 ml of THF, Hydroxybenzotriazole (HOBt) (1.5 eqv) was added and kept on stirring in ice water bath at 0°C. After 10 mins of stirring, DMAP (1 eqv) and a solution of DCC (1.93 eqv) in 5 ml of CHCl₃ was added to it and kept stirring. Finally, after 15 mins, K₂CO₃ (1.47 eqv) and PTSA-salt of 1-ω-Amino acid benzyl ester (1.2 eqv) was added and kept on stirring overnight at room temperature. The reaction mixture was then concentrated under reduced pressure and taken up in 30 ml CHCl₃ and washed sequentially with ice cold 1 (N) HCl (25 ml × 2), saturated brine (25 ml × 2). The organic layer was collected and dried over anhyd Na₂SO₄ and then purified by silicagel (60-120 mesh) column chromatography in EtOAc/Petroleum ether solvent system of required ratios (~40%-60%) to get the desired product.

**4a. Yield% = 77%.** δH (300 MHz, CDCl₃) : 0.857-0.918 (t, -NHCOCH₂CH₂(CH₂)₆CH₃ and >CHCH (CH₃)₂, 9H), 1.27 (bs, -NHCOCH₂CH₂(CH₂)₆CH₃, 12H), 1.58-1.65 (m, -NHCOCH₂CH₂(CH₂)₆CH₃, >CHCH (CH₃)₂3H), 2.11-2.24 (t, -NHCOCH₂CH₂(CH₂)₆CH₃, 2H), 2.59-2.62 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.5-3.63 (m, CONHCH₂CH₂COOCH₂Ph, 2H), 4.17-4.22 (t, >CHCH (CH₃)₂, 1H), 5.09-5.1 (s, -CONHCH₂CH₂COOCH₂Ph, 2H), 6.11-6.13 (d,- NHCOCH₂CH₂(CH₂)₆CH₃), 1H), 6.52 (s, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.26-7.35 (bs, aromatic, 5H). δC: 173.42, 172.56, 171.41, 135.58, 128.62, 128.4, 128.2, 77.46, 77.04, 76.61, 66.54, 58.35 36.73, 36.67, 35.00, 33.96, 31.9, 31.2, 29.63, 29.49, 29.33, 29.28, 25.74, 22.64, 19.12, 18.24, 14.07. ESI-MS (m/z) C₂H₄N₂O₄ (EXACT MASS= 432.3) 433.3(100%, [M + H]+), 455.3(49%, [M + Na]+).

**4b. Yield=73%.** δH (300 MHz, CDCl₃) : 0.87-0.91 (t, -NHCOCH₂CH₂(CH₂)₆CH₃ and >CHCH (CH₃)₂, 9H), 1.269 (bs, -NHCOCH₂CH₂(CH₂)₆CH₃, 16H), 1.63-1.74 (m, -NHCOCH₂CH₂(CH₂)₆CH₃, >CHCH (CH₃)₂3H), 2.11-2.2 (t, -NHCOCH₂CH₂(CH₂)₆CH₃, 2H), 2.58-2.62 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.5-3.63 (m, CONHCH₂CH₂COOCH₂Ph, 2H), 4.17-4.22 (t, >CHCH (CH₃)₂, 1H), 5.12 (s, -CONHCH₂CH₂COOCH₂Ph, 2H), 6.10-6.13 (d,- NHCOCH₂CH₂(CH₂)₆CH₃), 1H), 6.51(s, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.37 (bs, aromatic, 5H). δC: 173.2, 171.97, 171.35, 135.58, 128.62, 128.4, 128.3, 77.46, 77.04, 76.61, 66.61, 58.28 36.73, 34.9, 33.9, 31.8, 31.2, 29.6, 29.4, 29.32, 29.2, 25.74, 22.64, 19.12, 18.24, 14.09. ESI-MS (m/z) C₂H₄N₂O₄ (EXACT MASS= 460.33) 461.34(100%, [M + H]+), 483.32(34%, [M + Na]+).

**4c. Yield=76%.** δH (300 MHz, CDCl₃) : 1.1-1.25 (t, -NHCOCH₂CH₂(CH₂)₁₀CH₃ and >CHCH (CH₃)₂, 9H), 1.57 (bs, -NHCOCH₂CH₂(CH₂)₁₀CH₃, 20H), 1.87-2.03 (m, -NHCOCH₂CH₂(CH₂)₁₀CH₃, >CHCH (CH₃)₂3H), 2.3-2.4 (t, -NHCOCH₂CH₂(CH₂)₁₀CH₃, 2H), 2.79-2.81 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.49 (s, CONHCH₂CH₂COOCH₂Ph, 2H), 4.19-4.32 (t, >CHCH (CH₃)₂, 1H), 5.12 (s, -CONHCH₂CH₂COOCH₂Ph, 2H), 6.12-6.13 (t,- NHCOCH₂CH₂(CH₂)₁₀CH₃), 1H), 6.63-6.51(t, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.29-7.37 (m, aromatic, 5H). δC: 173.27, 172.93, 171.407, 135.6, 128.62, 128.4, 128.28, 77.46, 77.041, 76.61, 66.59, 58.256, 36.7, 34.976, 33.99, 33.69, 31.9, 31.2, 29.63, 29.49, 29.33, 29.28, 25.75, 25.54, 25.544, 24.865, 24.77, 22.67, 19.13, 18.22, 14.09. ESI-MS (m/z) C₂H₄N₂O₄ (EXACT MASS= 488.36) 489.369(100%, [M + H]+), 511.36(51%, [M + Na]+).

**4d. Yield=72%.** δH (300 MHz, CDCl₃) : 0.875-0.918 (m, -NHCOCH₂CH₂(CH₂)₁₂CH₃ and >CHCH (CH₃)₂, 9H), 1.27 (bs, -NHCOCH₂CH₂(CH₂)₁₂CH₃, 24H), 1.58-1.65 (m, -NHCOCH₂CH₂(CH₂)₁₂CH₃, 24H), 1.87-2.03 (m, -NHCOCH₂CH₂(CH₂)₁₂CH₃, >CHCH (CH₃)₂, 9H), 1.57 (bs, -NHCOCH₂CH₂(CH₂)₁₂CH₃, 20H), 1.87-2.03 (m, -NHCOCH₂CH₂(CH₂)₁₂CH₃, >CHCH (CH₃)₂3H), 2.3-2.4 (t, -NHCOCH₂CH₂(CH₂)₁₂CH₃, 2H), 2.79-2.81 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.49 (s, CONHCH₂CH₂COOCH₂Ph, 2H), 4.19-4.32 (t, >CHCH (CH₃)₂, 1H), 5.12 (s, -CONHCH₂CH₂COOCH₂Ph, 2H), 6.12-6.13 (t,- NHCOCH₂CH₂(CH₂)₁₂CH₃), 1H), 6.63-6.51(t, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.29-7.37 (m, aromatic, 5H). δC: 173.27, 172.93, 171.407, 135.6, 128.62, 128.4, 128.28, 77.46, 77.041, 76.61, 66.59, 58.256, 36.7, 34.976, 33.99, 33.69, 31.9, 31.2, 29.63, 29.49, 29.33, 29.28, 25.75, 25.54, 25.544, 24.865, 24.77, 22.67, 19.13, 18.22, 14.09. ESI-MS (m/z) C₂H₄N₂O₄ (EXACT MASS= 488.36) 489.369(100%, [M + H]+), 511.36(51%, [M + Na]+).
(CH₂)₂CH₃>CHCH (CH₃)₂, 3H), 2.11-2.24 (t, -NHCOCH₂CH₂(CH₂)₁₉CH₃, 2H), 2.59-2.62 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.5-3.6 (m, CONHCH₂CH₂COOCH₂Ph, 2H), 4.17-4.2 (t, >CHCH (CH₃)₂, 1H), 5.09-5.1 (s, -CONHCH₂CH₂COOCH₂Ph, 2H), 6.11-6.13 (d, -NHCOCH₂CH₂(CH₂)₁₉CH₃, 1H), 6.52 (s, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.26-7.35 (bs, aromatic, 5H). 

13C NMR (75 MHz, CDCl₃) : δc : 7.36, 172.4, 171.9, 135.6, 128.6, 128.3, 128.1, 77.46, 77.06, 76.61, 76.48, 74.27, 34.13, 33.39, 31.9, 31.17, 29.68, 29.65, 29.61, 29.5, 29.33, 29.26, 25.64, 24.81, 22.82, 22.66, 22.20, 14.08. ESI-MS (m/z) C₃₃H₅₂N₂O₄ (EXACT MASS = 516.37) 517.379 (100%, [M + H]+), 539.37 (20%, [M + Na]+).

4e. Yield: 69%. 1H NMR (300 MHz, CDCl₃) : δ : 0.85-0.89 (m, -NHCOCH₂CH₂ (CH₂)₁₉CH₃ and >CHCH (CH₃)₂, 9H), 1.24 (bs, -NHCOCH₂CH₂(CH₂)₁₉CH₃, 28H), 1.6 (s, -NHCOCH₂CH₂ (CH₂)₁₉CH₃ >CHCH (CH₃)₂, 3H), 2.14-2.19 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 2.56-2.60 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.5-3.54 (t, CONHCH₂CH₂COOCH₂Ph, 2H), 4.35-4.42 (t, >CHCH (CH₃)₂, 1H), 5.13 (s, -CONHCH₂CH₂COOCH₂Ph, 2H), 6.11-6.13 (d, -NHCOOCH₂CH₂ (CH₂)₁₉CH₃, 1H), 6.62 (s, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.35 (bs, aromatic, 5H). 13C NMR (75 MHz, CDCl₃) : δc : 173.24, 172.56, 135.62, 128.62, 128.4, 128.3, 77.44, 77.21, 76.6, 66.68, 58.35, 36.75, 34.97, 34.8, 34.13, 33.39, 31.9, 31.17, 29.68, 29.65, 29.61, 29.48, 29.34, 29.26, 25.73, 25.68, 22.68, 19.1, 18.2, 14.1. ESI-MS (m/z) C₃₃H₅₂N₂O₄ (EXACT MASS = 544.42) 545.429 (100%, [M + H]+), 567.4 (49%, [M + Na]+).

4f. Yield: 69%. 1H NMR (300 MHz, CDCl₃) : δ : 0.85-0.89 (m, -NHCOCH₂CH₂ (CH₂)₁₉CH₃ and >CHCH (CH₃)₂, 9H), 1.24 (bs, -NHCOCH₂CH₂(CH₂)₁₀CH₃, 28H), 1.58-1.65 (m, -NHCOCH₂CH₂ (CH₂)₁₀CH₃ >CHCH (CH₃)₂, 3H), 1.93-1.94 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 2.08-2.17 (t, -NHCOCH₂CH₂ (CH₂)₁₀CH₃, 2H), 2.55-2.59 (t, -CONHCH₂CH₂COOCH₂Ph, 2H), 3.44-3.59 (m, -CONHCH₂CH₂COOCH₂Ph, 2H), 4.37-4.39 (t, >CHCH (CH₃)₂, 1H), 5.132 (bs, -CONHCH₂CH₂ (CH₂)₁₀CH₃, 1H), 6.58 (s, -CONHCH₂CH₂COOCH₂Ph, 1H), 7.35 (bs, aromatic, 5H). 13C NMR (75 MHz, CDCl₃) : δc : 173.45, 173.26, 171.39, 135.75, 128.54, 128.32, 128.22, 77.45, 77.23, 77.03, 76.61, 66.44, 58.36, 38.92, 36.78, 36.70, 31.92, 31.76, 31.62, 30.95, 29.49, 29.35, 25.76, 24.61, 24.47, 22.68, 19.26, 14.11. ESI-MS (m/z) C₂₇H₄₄N₂O₄ (EXACT MASS = 502.38) 503.389 (100%, [M + H]+), 525.38 (68%, [M + Na]+), 541.3 (28%, [M + K]+).

4g. Yield: 69%. 1H NMR (300 MHz, CDCl₃) : δ : 0.84-0.85 (m, -NHCOCH₂CH₂ (CH₂)₁₉CH₃ and >CHCH (CH₃)₂, 9H), 1.25 (bs, -NHCOCH₂CH₂(CH₂)₁₀CH₃, -CONHCH₂CH₂(CH₂)₁₀CH₃, 28H), 1.6 (s, -NHCOCH₂CH₂(CH₂)₁₀CH₃ >CHCH (CH₃)₂, 3H), 1.78-1.78 (d, -CONHCH₂CH₂(CH₂)₁₀CH₃, 2H), 2.08-2.2 (t, -NHCOCH₂CH₂ (CH₂)₁₀CH₃, 2H), 2.56-2.60 (t, -CONHCH₂CH₂ (CH₂)₁₀CH₃, 2H), 3.44-3.58 (m, -CONHCH₂CH₂ (CH₂)₁₀CH₃, 2H), 4.17-4.22 (t, >CHCH (CH₃)₂, 1H), 5.09-5.13 (s, -CONH(CH₂)₁₀COOCH₂Ph, 2H), 6.01-6.04 (d, -NHCOCH₂CH₂ (CH₂)₁₀CH₃, 1H), 6.35 (s, -CONH(CH₂)₁₀COOCH₂Ph, 1H), 7.35 (bs, aromatic, 5H). 13C NMR (75 MHz, CDCl₃) : δc : 173.27, 172.93, 171.407, 135.58, 128.6, 128.4, 128.28, 77.46, 77.04, 76.61, 66.59, 58.256, 40.2, 36.73, 36.67, 35.00, 33.1, 31.84, 31.19, 29.63, 29.49, 29.33, 29.28, 25.74, 25.54, 22.64, 18.212, 17.124, 14.102. ESI-MS (m/z) C₂₇H₄₄N₂O₄ (EXACT MASS = 530.41) 531.419 (100%, [M + H]+), 553.421 (62%, [M + Na]+).

4h. Yield: 69%. 1H NMR (300 MHz, CDCl₃) : δ : 0.846-0.934 (m, -NHCOCH₂CH₂ (CH₂)₁₉CH₃ and >CHCH (CH₃)₂, 9H), 1.233-1.280 (m, -NHCOCH₂CH₂(CH₂)₁₀CH₃, -CONH(CH₂)₁₀CH₃, 28H), 1.476-1.495 (d, -NHCOCH₂CH₂(CH₂)₁₀CH₃, -CONHCH₂CH₂ (CH₂)₁₀CH₃, 2H), 1.970-2.0 (m, >CHCH (CH₃)₂, 1H), 2.130-2.243 (m, -CONHCH₂CH₂ (CH₂)₁₀CH₃, 2H), 2.3-2.36 (m, -NHCOCH₂CH₂(CH₂)₁₀CH₃, 2H), 3.2-3.266 (m, -CONHCH₂CH₂COOCH₂Ph, 2H), 4.084-4.25
(m, >CHCH (CH₃)₂, 1H), 5.11 (s, -CONH(CH₂)₁₀COOCH₂Ph, 2H), 6.625-6.719 (m, -NHCO(CH₂)₆CH₃), -CONH(CH₂)₁₀COOCH₂Ph, 2H), 7.35 (s, aromatic, 5H). ¹³C NMR (75 MHz, CDCl₃) : δ C: 173.42, 172.56, 171.41, 135.58, 128.62, 128.4, 128.2, 77.46, 77.04₁, 76.61, 66.54, 58.35, 40.3, 36.73, 36.67, 35.00, 33.1, 31.84, 31.19, 29.7, 29.7, 29.43, 29.32, 29.24, 28.7, 25.74, 22.64, 17.4, 17.1, 14.1. ESI-MS (m/z) C₂₇H₄₄N₂O₄ (EXACT MASS = 516.37) 517.379(100%, [M + H]+), 539.39(58%, [M + Na]+), 555.329(18%, [M + K]+).

¹H NMR spectra of gelator 4a.

¹H NMR spectra of gelator 4b.
$^1$HNMR spectra of gelator 4c.

$^1$HNMR spectra of gelator 4d.
HNMR spectra of gelator 4e.
MR spectra of gelator \(4g\).

NMR spectra of gelator \(4h\).
$^{13}$C NMR spectra of gelator 4a.

$^{13}$C NMR spectra of gelator 4b.
$^{13}$C NMR spectra of gelator **4c**.

$^{13}$C NMR spectra of gelator **4d**.
$^{13}$C NMR spectra of gelator 4e.

$^{13}$C NMR spectra of gelator 4f.
$^{13}$C NMR spectra of gelator 4g.

$^{13}$C NMR spectra of gelator 4h.
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© DCB - Dichlorobenzene | PE - Petroleum ether | EG - Ethylene Glycol
Rheology:

Figure S1. Strain amplitude sweep experiment ($\omega$ at 10rads/sec) for (4b) in acetonitrile.

Figure S2. Strain amplitude sweep experiment ($\omega$ at 10rads/sec) for (4d) in acetonitrile.

Figure S3. Strain amplitude sweep experiment ($\omega$ at 10rads/sec) for (4e) in acetonitrile.
Continuation of Rheology….

Figure S4. Strain amplitude sweep experiment (ω at 10rads/sec) for (4f) in acetonitrile.

Thermal stability

Figure S5. Temperature sweep experiment for (4c) in acetonitrile

Figure S6. Temperature sweep experiment for (4f) in acetonitrile
Continuation of Thermal Stability…..

Figure S7. Temperature sweep experiment for (4g) in acetonitrile

Figure S8. $T_{gel}$ (°C) versus concentration (mnoles) plots for gelators of (a) SET-II and (b) SET-III in Acetonitrile.
XRD:

Figure S9. XRD profile of xerogels (intensity vs. 2θ) for 4a in acetonitrile and its corresponding plot of $d^{-1}$ vs. $\sqrt{(h^2+l^2+k^2)}$.

Figure S10. XRD profile of xerogels (intensity vs. 2θ) for 4b in acetonitrile and its corresponding plot of $d^{-1}$ vs. $\sqrt{(h^2+l^2+k^2)}$. 
Continuation of XRD….

Figure S11. XRD profile of xerogels (intensity vs. 2θ) for 4c in acetonitrile and its corresponding plot of d⁻¹ vs. \(\sqrt{h^2+l^2+k^2}\).

Figure S12. XRD profile of xerogels (intensity vs. 2θ) for 4e in acetonitrile and its corresponding plot of d⁻¹ vs. \(\sqrt{h^2+l^2+k^2}\).
Continuation of XRD….

Figure S13. XRD profile of xerogels (intensity vs. 2θ) for 4g in 80%(v/v) Ethanol.H₂O and its corresponding plot of d⁻¹ vs. √(h²+l²+k²) (at inset).

Figure S14. XRD profile of xerogels (intensity vs. 2θ) for 4h in acetonitrile and its corresponding plot of d⁻¹ vs. √(h²+l²+k²).

Figure S14. XRD profile of xerogels (intensity vs. 2θ) for 4g in anisole and its corresponding plot of d⁻¹ vs. √(h²+l²+k²)
Continuation of XRD….

Figure S15. XRD profile of xerogels (intensity vs. 2θ) for 4c in 80% aq. EtOH and its corresponding plot of d⁻¹ vs. \( \sqrt{h^2+l^2+k^2} \).

Figure S16. XRD profile of xerogels (intensity vs. 2θ) for 4c in Anisole and its corresponding plot of d⁻¹ vs. \( \sqrt{h^2+l^2+k^2} \).

Figure S17. XRD profile of xerogels (intensity vs. 2θ) for 4c in EtOH and its corresponding plot of d⁻¹ vs. \( \sqrt{h^2+l^2+k^2} \).

Solvent effect:

Figure S18. \( E_{T30} \) versus \( T_g \) plot of gelator 4a, 4b, 4c, 4d and 4e in different solvents.
Figure S19. 3D plots of Hansen and Kamlet-Taft parameters at different solvent for 4a-4g gelators.

Figure S20. Comparison of 3D plots of Kamlet-Taft parameters at different solvents for 4d-4h gelators.
Continuation of Solvent effect….

Figure S21. Plots of δₚ vs δₜₜ, δₐ vs δₜₜ and δₚ vs δₜ (HSP) for gelator 4h [gelation is found in solvents with lower δₜ values, i.e., solvents with lower H-bonding capability].

Figure S22. Plots of α vs β, α vs π*, and π* vs β. (KT parameters) for gelator 4h.

Figure S23. Plots of δₜ vs δₜ for gelators 4a-4g.
Continuation of Solvent effect….

Figure S24. Teas plot of Hansen Parameters for gelators of Set-I and Set –II in solvents of gelation.

Figure S25. Teas plot of Hansen Parameters for 4h in solvents of gelation.
Figure S26. CD spectra of 4f at different concentrations.

Figure S27. CD spectra of 4f at different concentrations.

Figure S28. CD spectra of 4f at different concentrations.
Continuation of CD.....

Figure S29. CD spectra of 4f at different concentrations.

Figure S30. CD spectra of 4f at different concentrations.
FT-IR spectra:

Figure S31. IR spectra of gelator 4d in acetonitrile in gel state.

Figure S32. IR spectra of gelator 4h in acetonitrile in gel state.
Figure S33. IR spectra of gelator 4a in acetonitrile in gel state.

Figure S34. IR spectra of gelator 4b in acetonitrile in gel state.
Figure S35. IR spectra of gelator 4c in acetonitrile in gel state.

Figure S36. IR spectra of gelator 4e in acetonitrile in gel state.
Figure S37. IR spectra of gelator 4f in acetonitrile in gel state.

Figure S38. IR spectra of gelator 4g in acetonitrile in gel state.