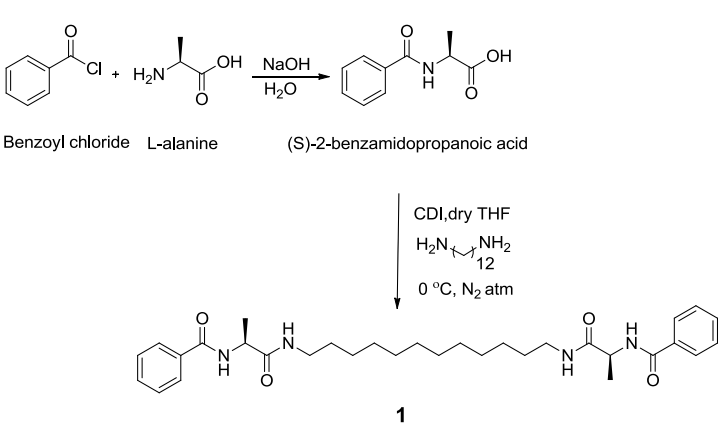
# Supporting Information.

Includes synthesis, $^{1}H$, $^{13}C$, DEPT NMR of **1**, MGC data, FTIR, CD, DSC and $^{1}H$-$^{1}H$ COSY of the gel before and after sonication, proposed model of assembly, Jobs plot and individual binding constant vs *p* plots for the $^{1}H$ NMR titration of **1** with TBAF and TBACl and other studies.




**Scheme S1**: Synthesis of gelator

**Synthesis of the bolaaphiphilic peptide (1).** (*S*)-2-benzamidopropanoic acid[1] (2.2 eq, 2.12 g, 11 mmol) and CDI (2.2 eq, 1.78 g, 11 mmol) were taken in dry THF and stirred for 5 min. The 1,12-diamino-dodecane (1 eq, 1 g, 5 mmol) in dry THF was added dropwise at 0 ˚C to the activated acid solution, and the reaction mixture was stirred for 1.5 h under $N_2$ atmosphere. The reaction was monitored by TLC ($CHCl_3$/MeOH 9.5:0.5 v/v). THF was removed from the reaction mixture by vacuum and 50 mL of $CHCl_3$/isopropanol (9:1) was added to it. The organic layer was washed with dil. HCl and sat. $NaHCO_3$, and dried over anhyd. sodium sulphate. The solvent was then removed under rotary evaporation to yield the crude **1**, which was purified by column chromatography (stationary phase was silica 100-200 mesh and eluent was $CHCl_3$/MeOH 9:1 v/v) to obtain **1** as a white solid (2.5 g, 90%). m.p. 160 ˚C. FTIR (KBr pellet, $cm^{-1}$): 1637 (amide, C=O str.), 1535 (N-H bend), 3305 (N-H str.). $^{1}H$ NMR (400 MHz, DMSO-d6, ppm): δ= 1.12-1.24 (16H, $(CH_2)_4$), 1.25-1.31 (6H, $CH_3$),1.32-1.40 (4H, NH-$CH_2$-**$CH_2$**), 3.29 (4H, NH-**$CH_2$**), 4.38-4.41 (2H, C-**H**), 7.40-7.87(10H, Ph-H), 7.80-7.82 (N**H**$_b$), 8.37-8.39 (N**H**$_a$). $^{13}C$ NMR (400 MHz, DMSO) δ= 172.56, 166.41, 134.60, 131.69, 128.59, 127.96, 49.43, 38.95, 29.47, 29.44, 29.20, 26.74, 18.58. $C_{32}H_{46}N_4O_4$+$Na^+$ calc: 573.3411, found: 573.3405.

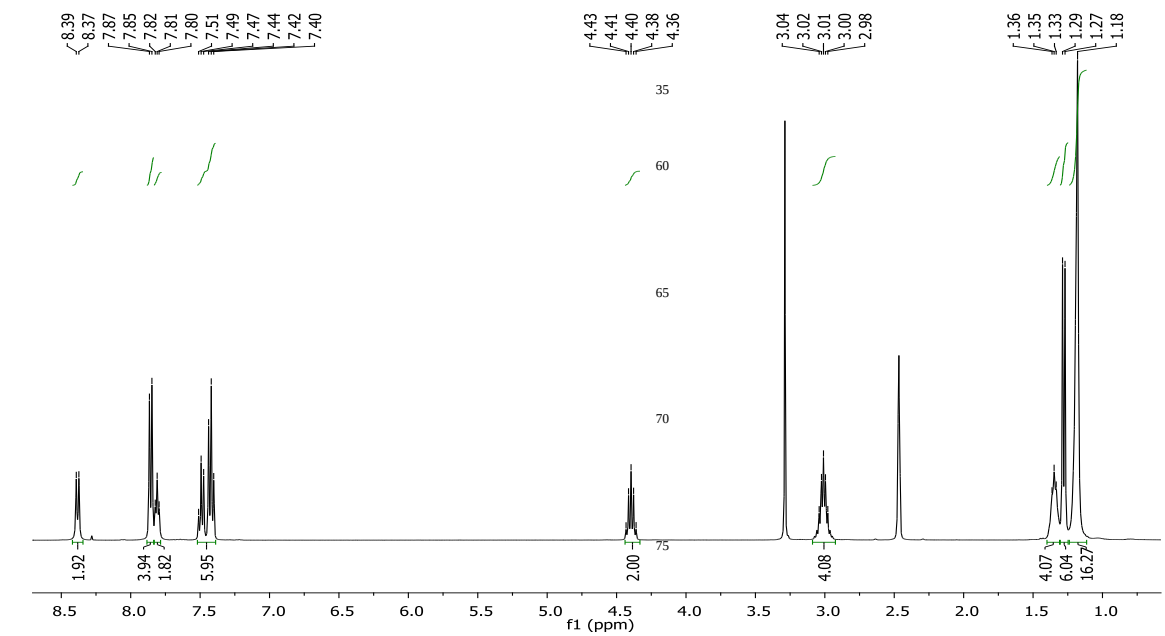



**Fig. S1**. $^{1}H$ NMR (400 MHz) spectra of compound **1** in DMSO-d6.

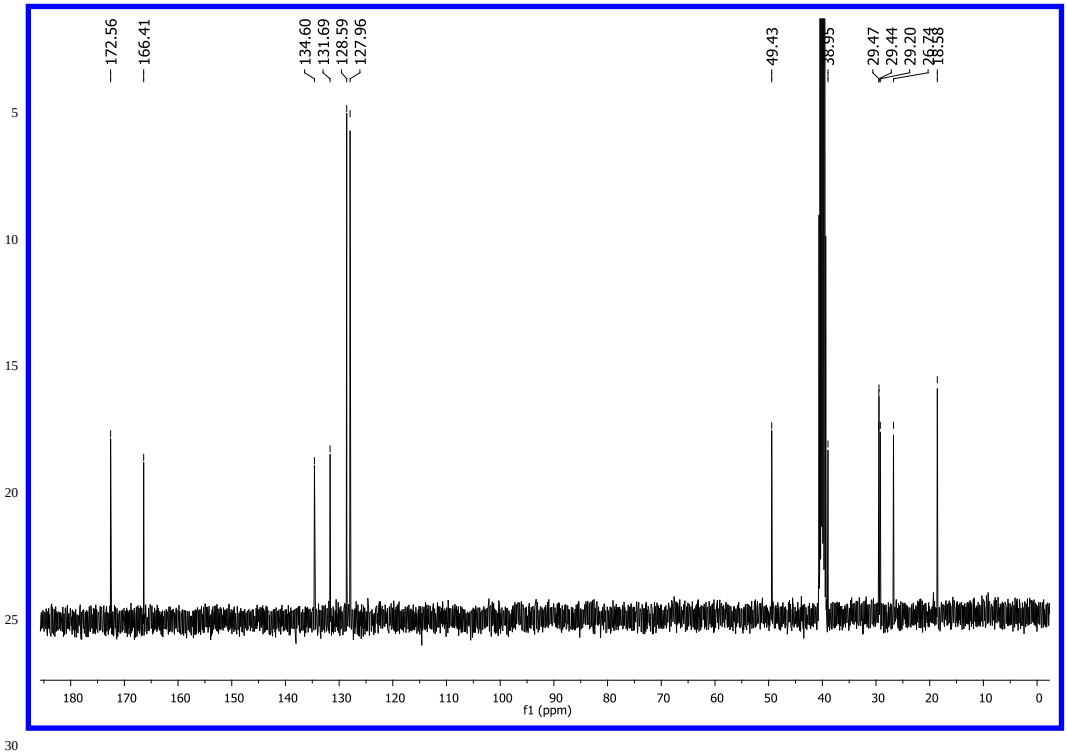



**Fig. S2**. $^{13}$C NMR (100 MHz) spectra of **1** in DMSO-d6.

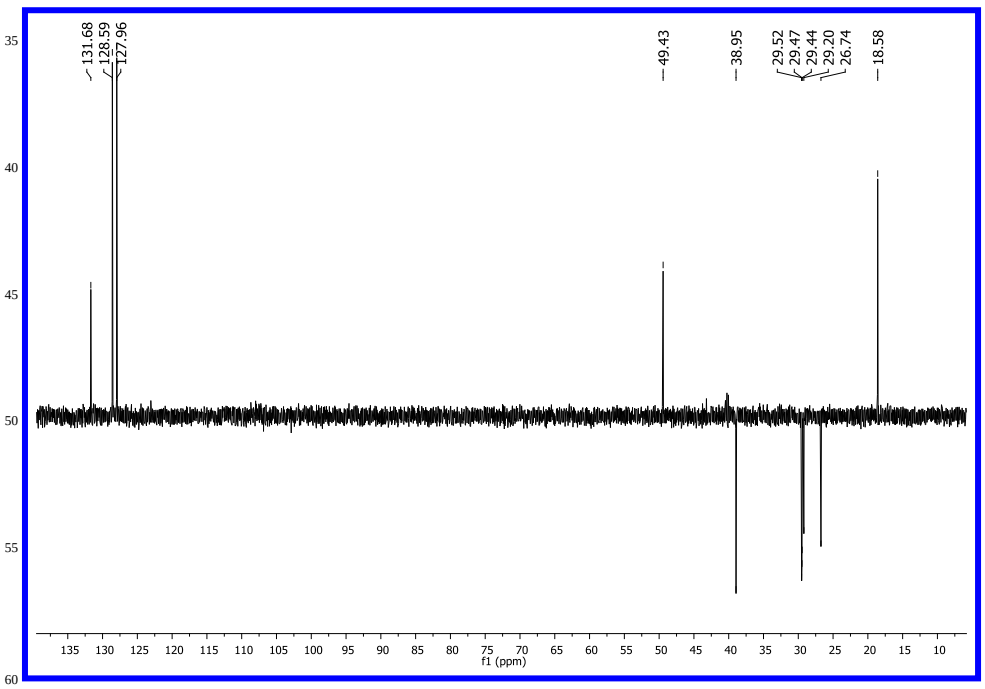



**Fig. S3**. DEPT (100 MHz) spectra of **1** in DMSO-d6.

| Solvent | MGC (%w/v) | $T_m$ (˚C) |
|---|---|---|
| THF | 1.0 | 45±3 |
| 1,4-Dioxane | 1.5 | 50±2 |

**Table S1**. MGC values and melting temperatures ($T_m$) at MGC

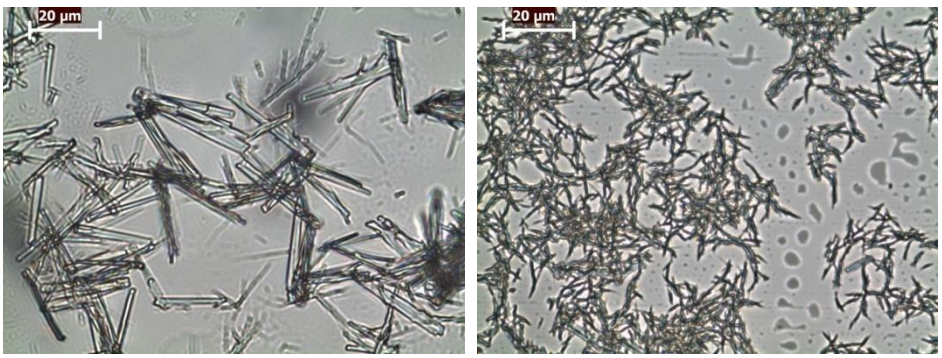



**Fig. S4**. Optical microscopy images (100x magnification) of: a) the microcrystals of **1**; b) short fibrils formed after 3 min sonication of the microcrystals

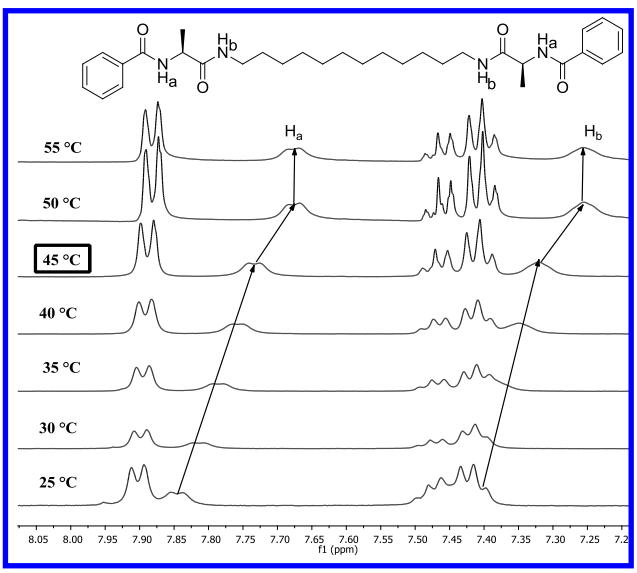



**Fig. S5**. Variable temperature $^1$H NMR of the sonication-induced assembly in THF-d8. The physical gel-to-sol transition temperature ($T_m$) around 45 °C is highlighted.

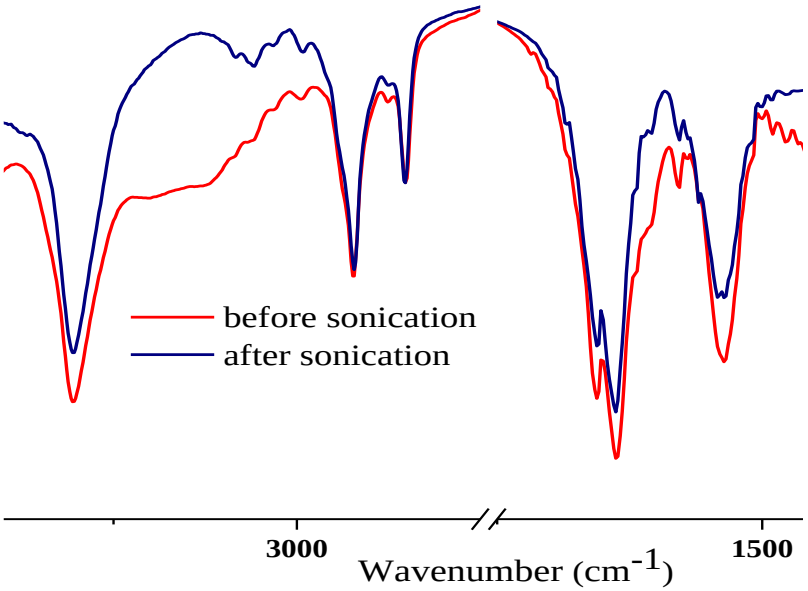



**Fig. S6**. FTIR spectra of **1** recorded as KBr pellets. Dried microcrystals before sonication (red line), and dried fibrils formed after sonication (blue line).

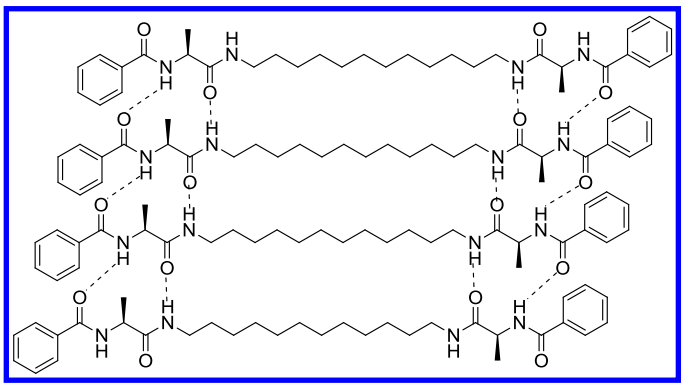


**Fig. S7**. Proposed model for the assembly of **1** by intermolecular hydrogen bonding interactions between the amide groups.

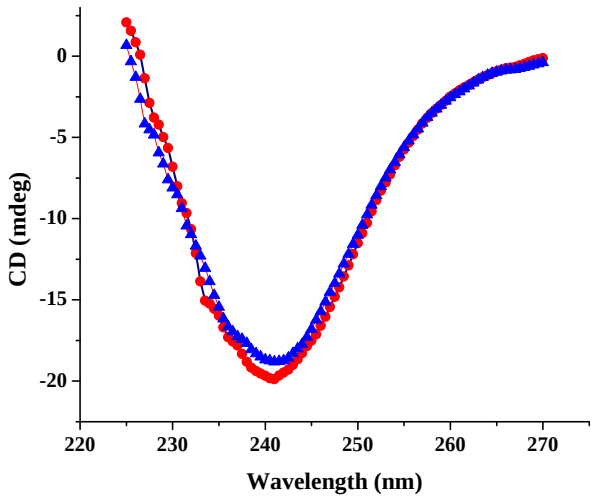



**Fig. S8**. CD spectra of **1** recorded as a solution in THF (0.75 mg/mL) at 25 ˚C. Before sonication (red circles), and after sonication (blue triangles).

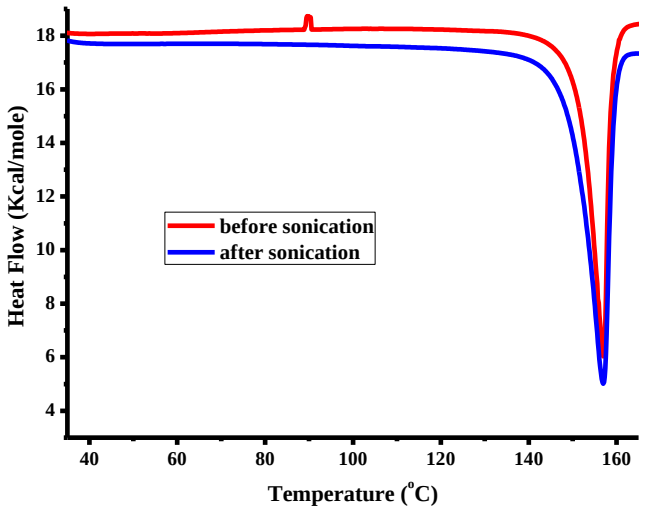



**Fig. S9**. DSC profiles of **1**, as dried microcrystals before sonication (red line), and as dried fibrils formed upon sonication (blue line). 4 mg of pre-dried sample was heated at the scan rate of 1°C/min.

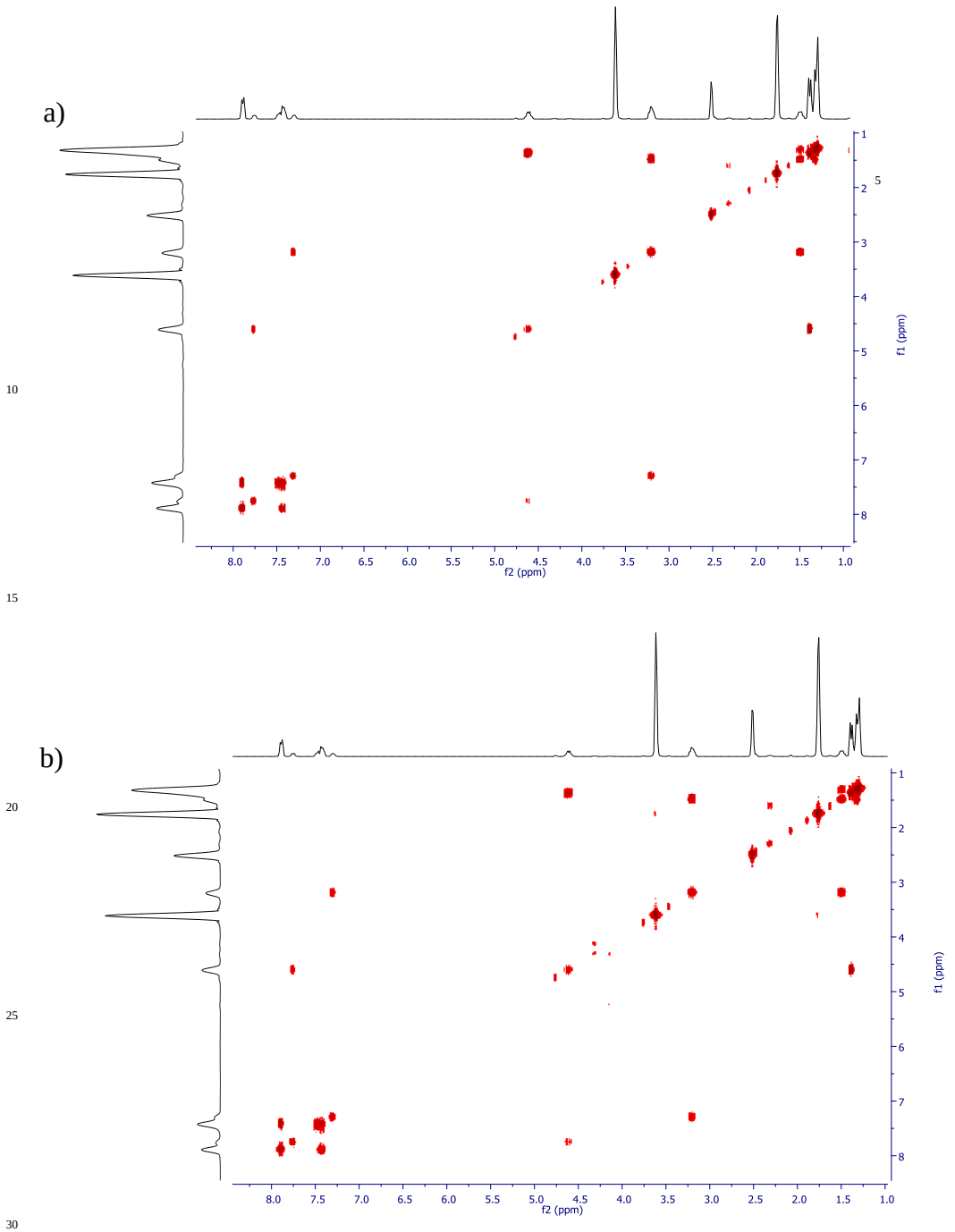



**Fig. S10**. $^{1}$H-$^{1}$H COSY spectra of **1** (5.5 mM) in THF-d8. a) before sonication; b) after sonication.

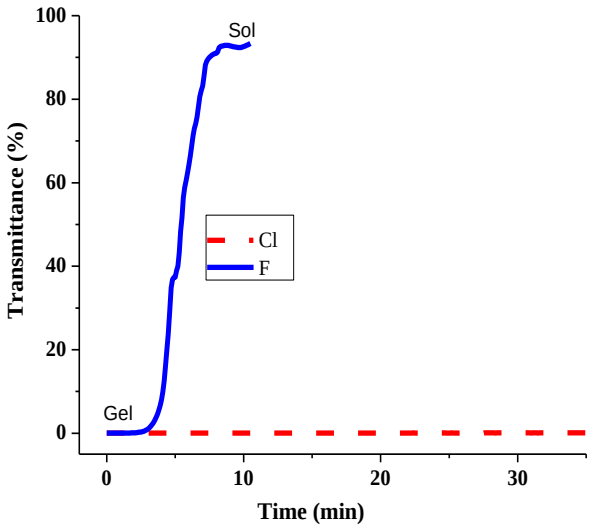



**Fig. S11**. Relative rates of anion-induced disassembly of nanofibers (prepared through sonication of **1** at MGC in THF) by $F^-$ or $Cl^-$ ions (4 eqv), as evaluated from the increase in %T at 600 nm.

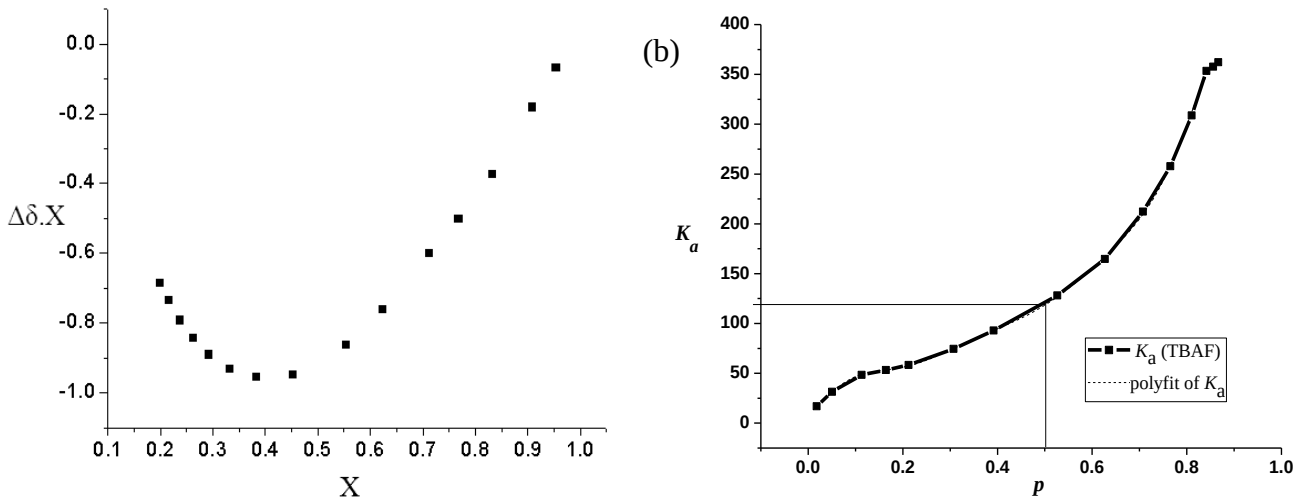



**Fig. S12**. (a) Jobs plot for the chemical shift of $NH_a$ proton during the titration of TBAF with **1**. X = mole-fraction of **1**, Δδ = change in the chemical shift of $NH_a$ proton. (b) $K_a$ vs. *p* for the $NH_a$ proton of **1** during NMR titration with TBAF; *p* is the probability of complexation (data fitted in polyfit using Origin™ 8.0 ($R^2$=0.999)).

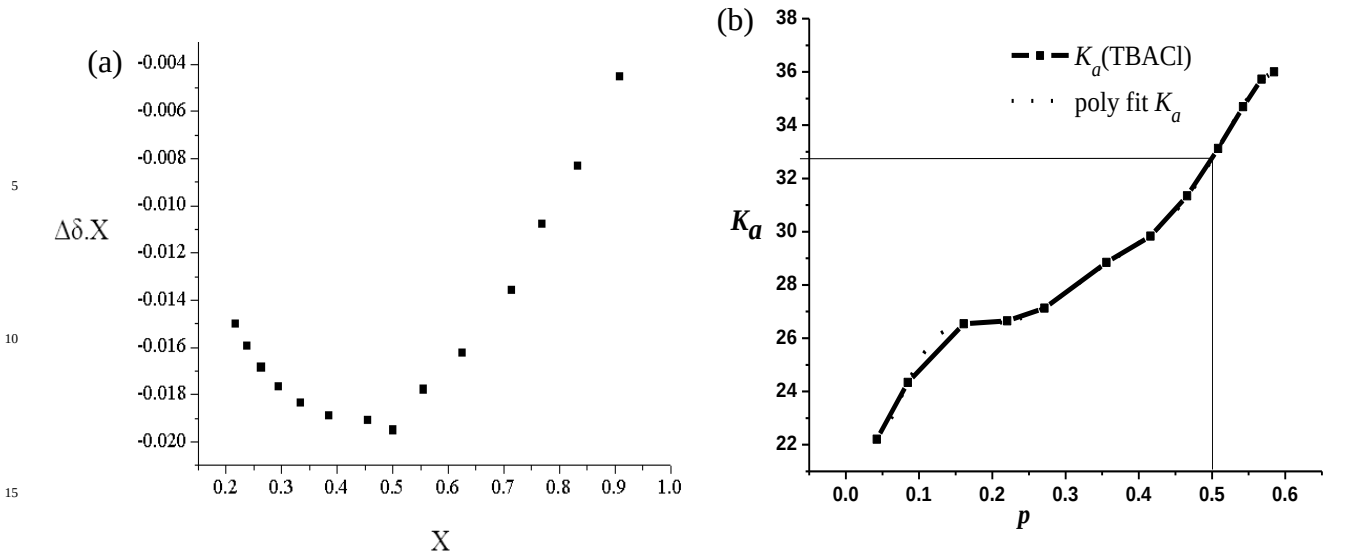



**Fig. S13**. (a) Jobs plot for the chemical shift of $NH_a$ proton during titration of TBACl with **1**. X = mole-fraction of **1**, Δδ = change in the chemical shift of $NH_a$ proton of **1**. (b) $K_a$ vs. *p* for the $NH_a$ proton of **1** during NMR titration with TBACl; *p* is the probability of complexation (data fitted in polyfit using Origin™ 8.0 ($R^2$=0.999)).

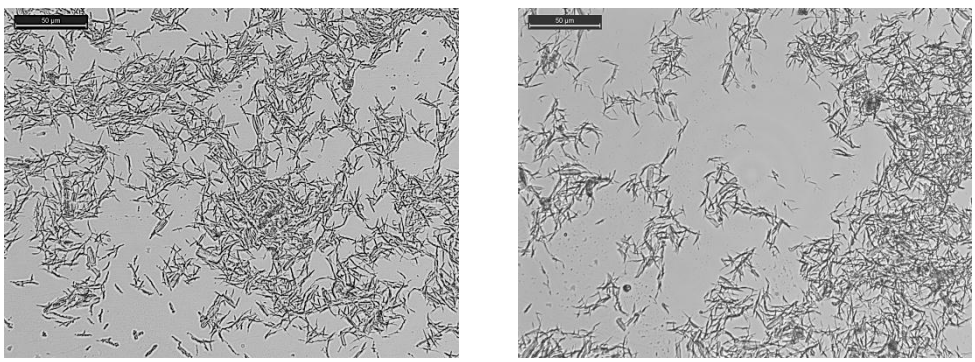


**Fig S14:** Optical microscopy images (40x magnification) of the regenerated gel exhibiting fibrous architecture similar to the pristine gel (scale 50 µm).

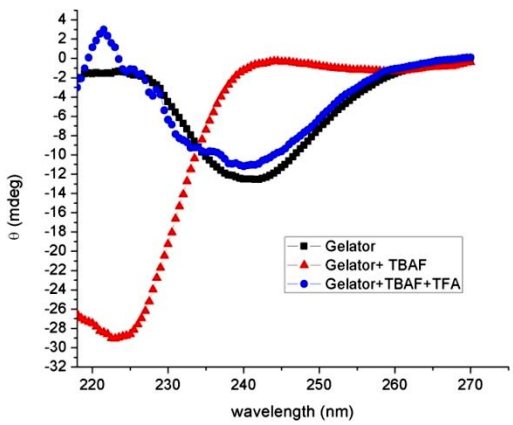



**Fig S15:** CD profiles of TFA regenerated gel showing the regeneration of the CD signal similar to the pristine gelator.

(1) A.I. Vogel, A.R. Tatchell, B.S. Furnis, A.J. Hannaford, P.W.G. Smith, in *Vogel's Textbook of Practical Organic Chemistry*, (5th ed.) Longman Group Ltd., London **1989** pp. 815.