Electronic Supplementary Material (ESI) for Chemical Communications

Supporting Information

Cooperative Self-Assembly of Linear Organogelators. Amplification of Chirality and Crystal Growth of Pharmaceutical Ingredients

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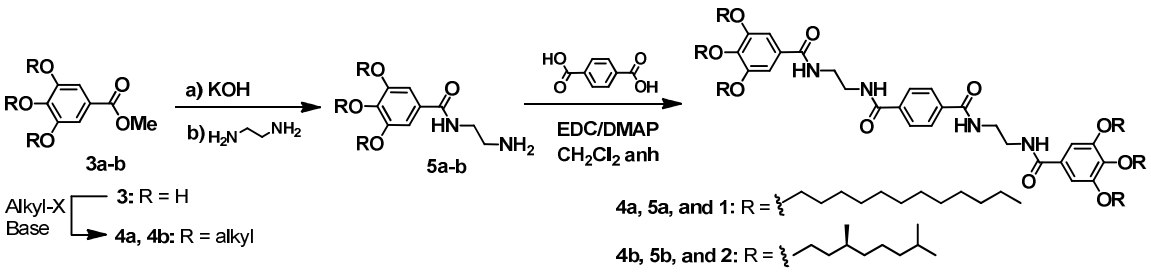
3. Supplementary Figures and Tables



Scheme S1. Synthesis of organogelators **1** and **2**.

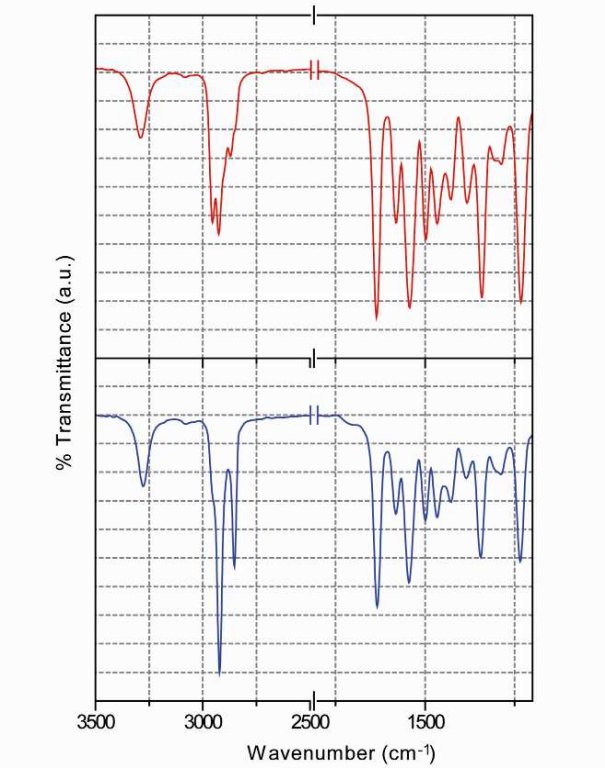


Figure S1. Partial FTIR spectra (film) of organogelators **1** and **2**.

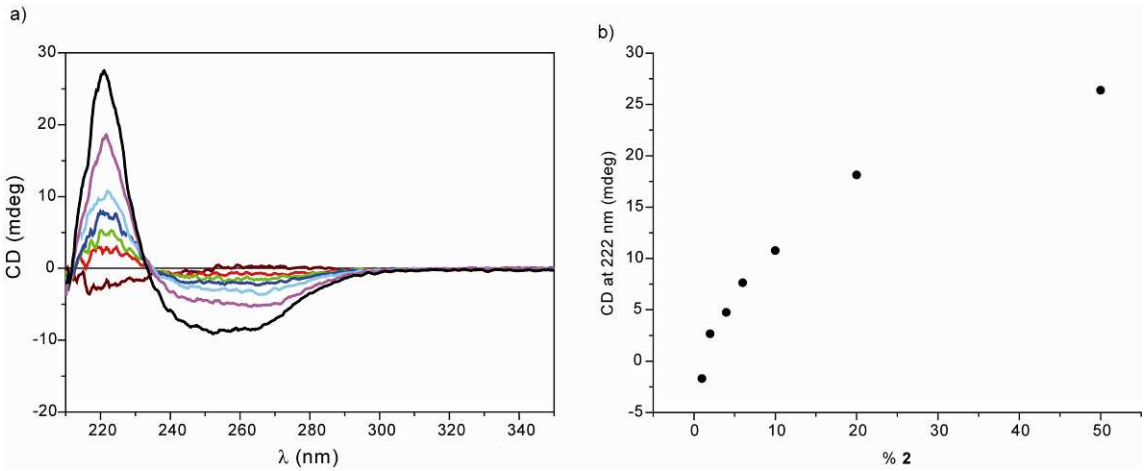


Figure S2. CD spectra (a) and amplification of chirality experiments (b) of achiral solution of **1** (298 K, MCH, 1×10^{-5} M) upon addition of increasing amounts of **2**.

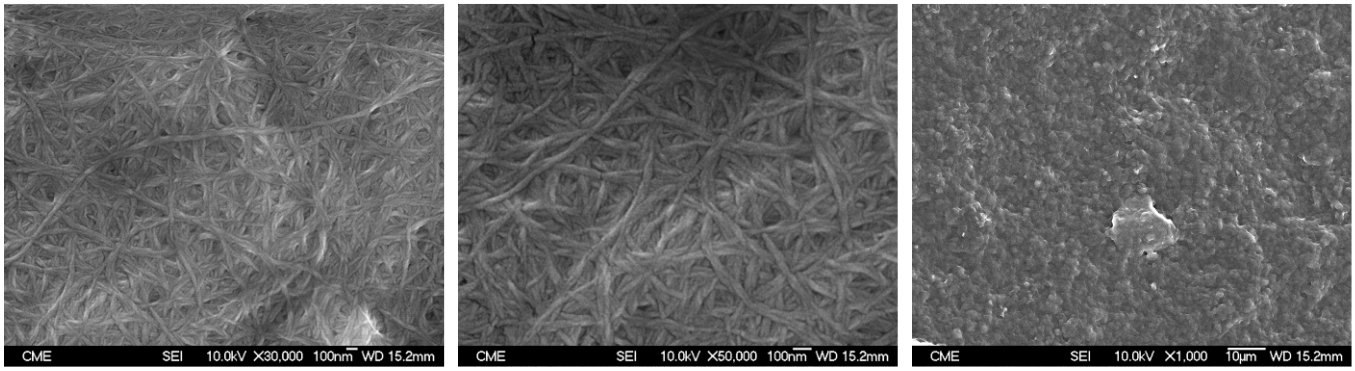


Figure S3. SEM images of the fibrillar network constitutive of the supramolecular gel of **1** (1 wt%) (left and middle) and **2** (1 wt%, right) in toluene.

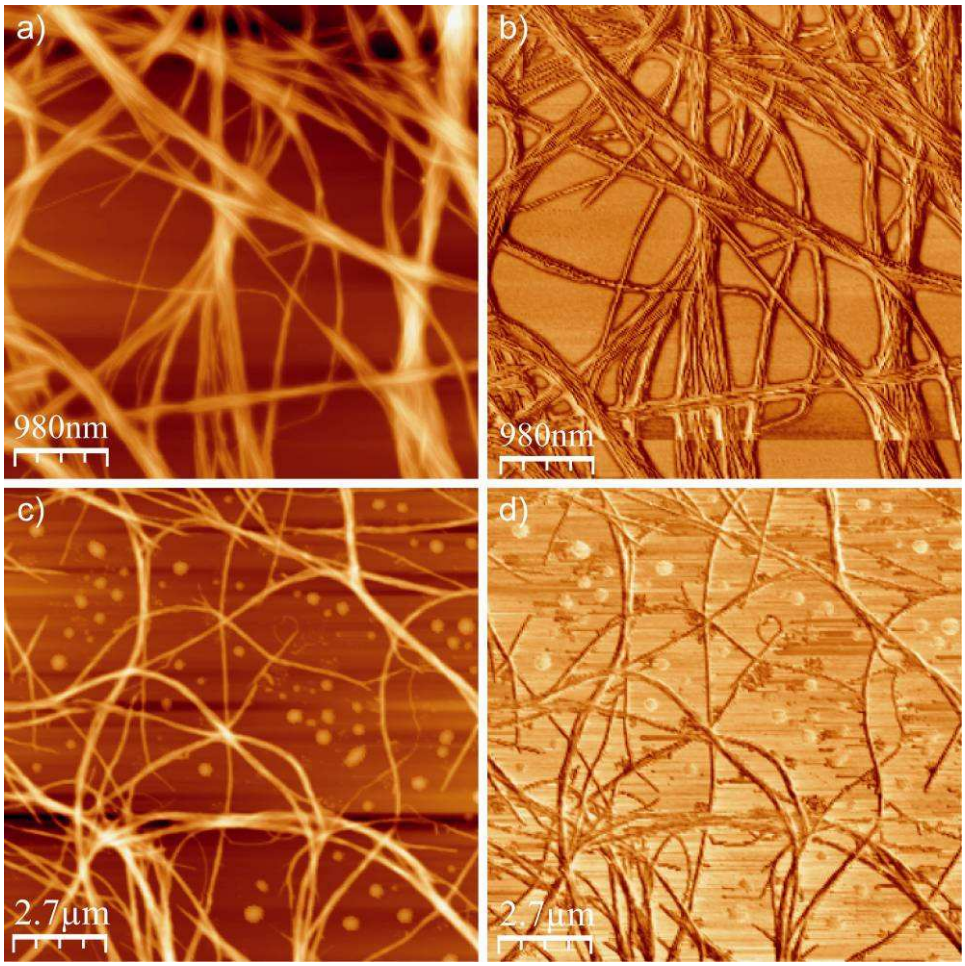


Figure S4. AFM images (height, left, and phase, right) of the fibres constitutive of the supramolecular gel of **1** (1 x 10^{-4} M, toluene) onto HOPG (a, b; z scale = 200 nm), and onto mica (c, d; z scale = 150 nm).

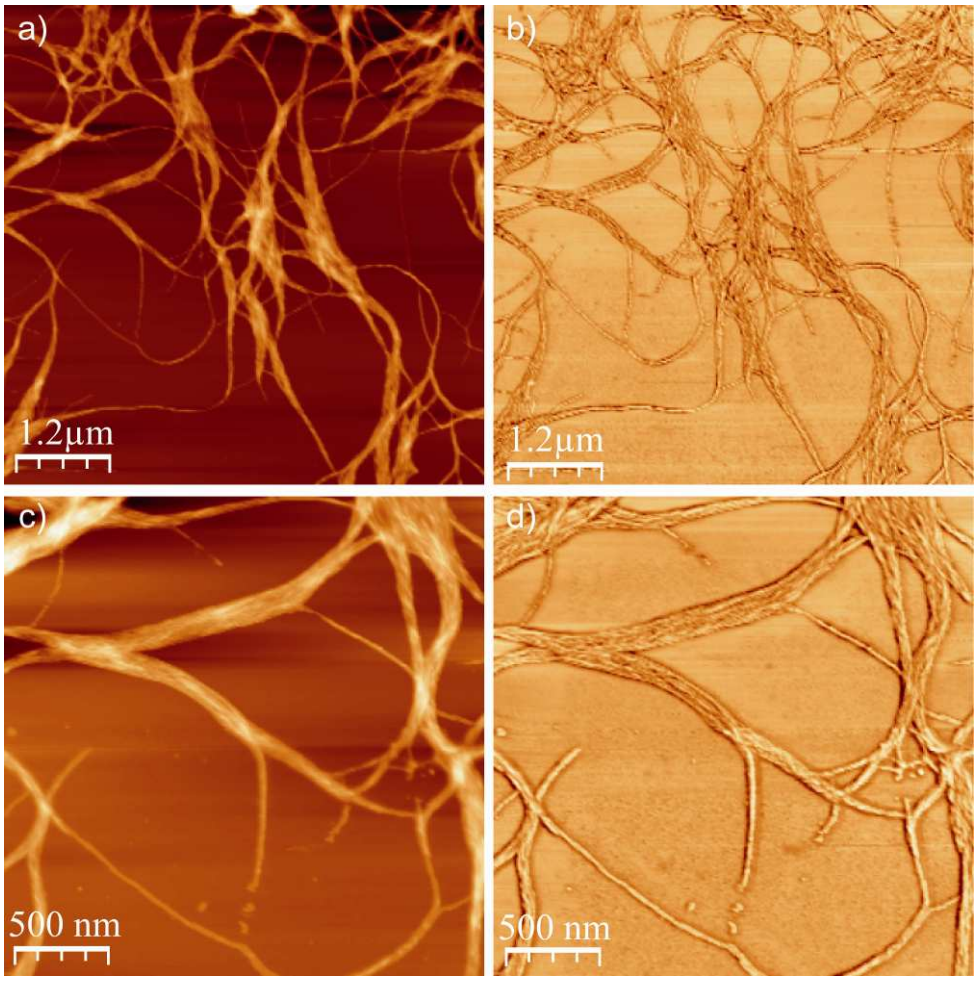


Figure S5. AFM images (height, left, and phase, right) of the fibres constitutive of the supramolecular gel of **2** (1 x 10^{-4} M, toluene, HOPG) at different magnifications.

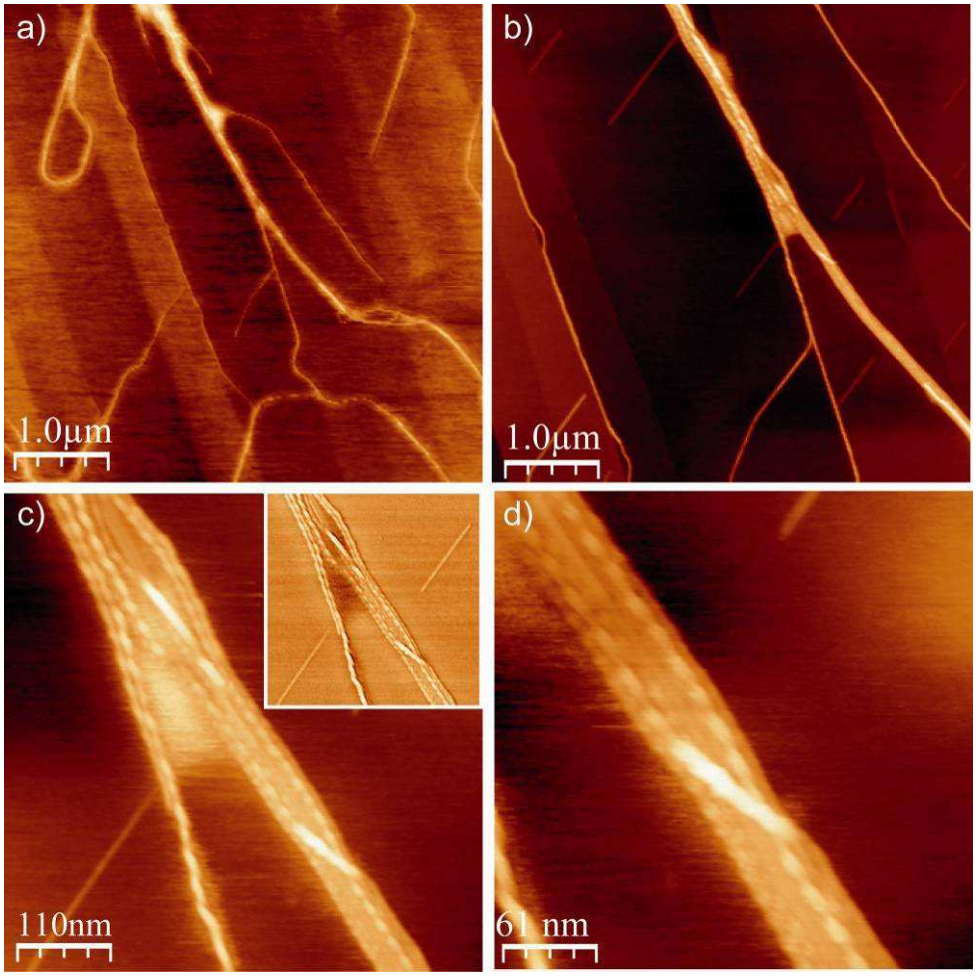


Figure S6. AFM images of the fibres constitutive of the supramolecular gel of **2** (1 x 10^{-5} M, toluene, HOPG) at different magnifications. The inset in (c) corresponds to the phase image in (c).

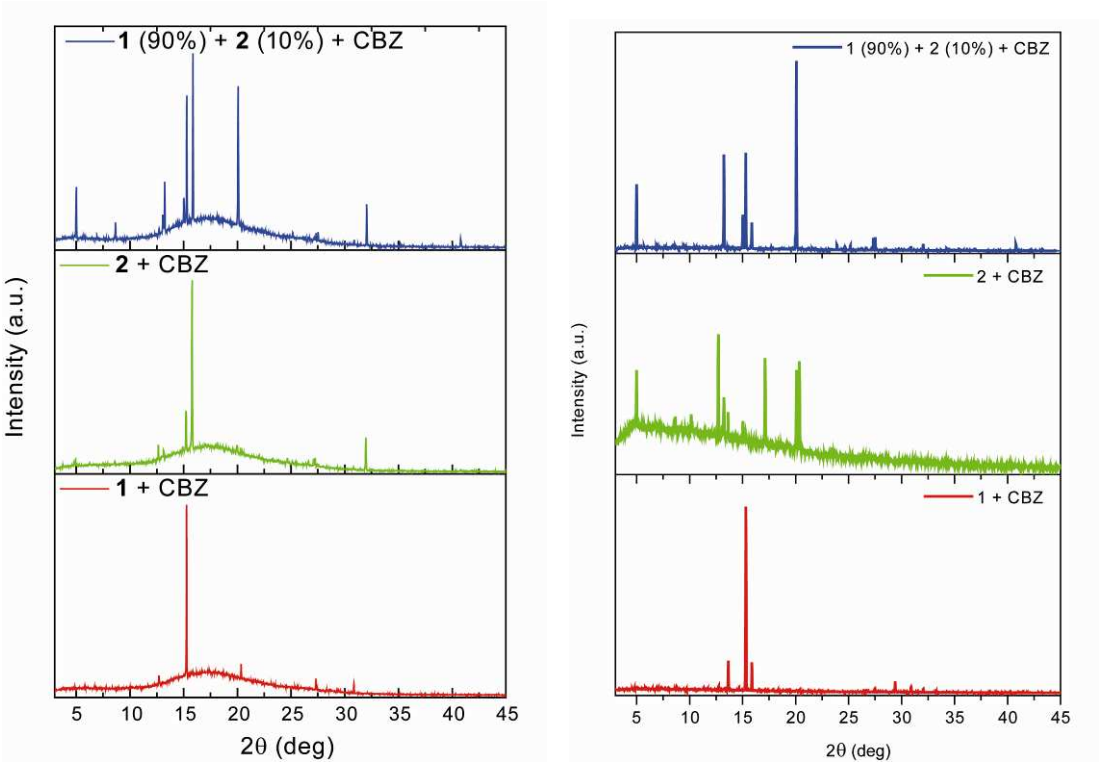


Figure S7. X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CBZ inside the toluene gels of **1** (bottom), **2** (middle), and the 9/1 mixture of **1** and **2** (top). The left panel shows the X-ray diffractograms of CBZ crystals obtained inside gel and before washing with toluene. The right panel shows the X-ray diffractograms of CBZ crystals after washing with toluene

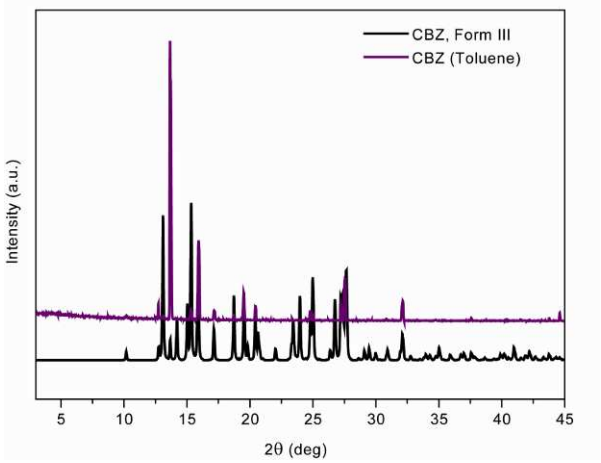


Figure S8. Comparison of the experimental XRD pattern of the crystals of CBZ obtained in toluene (red) with the calculated pattern for CSD entry CBMZPN01 (Reboul, J. P.; Cristau, B.; Soyfer, J. C.; Astier, J. P. *Acta Crystallogr. B,* **1981,** *37*, 1844-1848) generated using X'Pert HighScore Plus software (black).

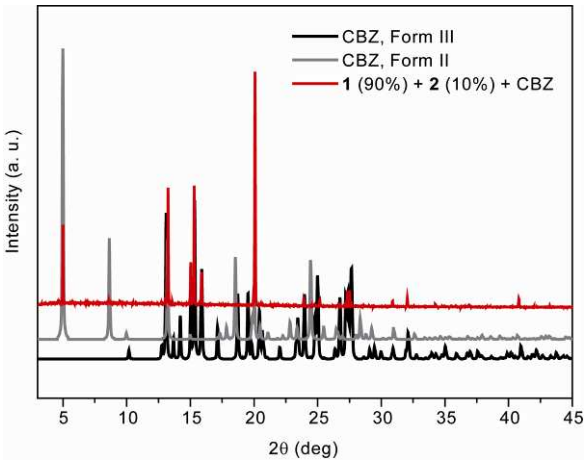


Figure S9. Comparison of the experimental XRD pattern of the crystals of CBZ obtained in the 9/1 mixture of organogelators **1** and **2** (red) with the calculated patterns for CSD entries CBMZPN01 (black; Reboul, J. P.; Cristau, B.; Soyfer, J. C.; Astier, J. P. *Acta Crystallogr. B,* **1981,** *37*, 1844-1848), and CBMZPN03 (grey; Lowes, M. M. J.; Cairo, M. R.; Lotter, A. P.; van der Watt, J. G. *J. Pharm. Sci.* **1987**, *76*, 744-752) generated using X'Pert HighScore Plus software.

Table S1. Polymorphic outcome for the crystallization of CBZ in solutions of achiral **1** or chiral **2** in toluene as solvent.[a]

Concentration (M)	Compound 1	Compound 2
1×10^{-6}	*II+III*	*II+III*
1×10^{-5}	*III*	*II+III*
$1 \times 10^{-3(b)}$	*III*	*II+III*
$\sim 5 \times 10^{-3(c)}$	*III*	*II+III*
$1 \times 10^{-2(d)}$	*III*	*III*

[a] Crystallization performed at a concentration of 1 wt% of CBZ.[(b,c)] This molarity corresponds to concentrations of 0.5 wt% (b), 1 wt% (c) and 2 wt% (d), respectively, and the organogel is formed.

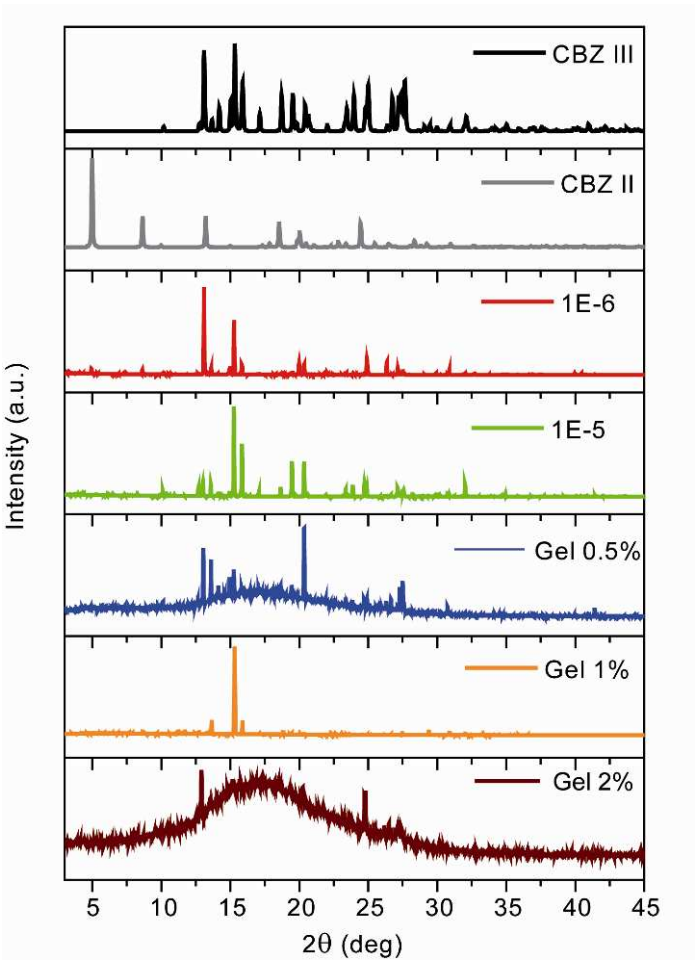


Figure S10. Comparison of the experimental XRD pattern of the crystals of CBZ obtained in the presence of organogelator **1** at different concentrations with the calculated pattern for CSD entry CBMZPN01 (black, Reboul, J. P.; Cristau, B.; Soyfer, J. C.; Astier, J. P. *Acta Crystallogr. B,* **1981,** *37*, 1844-1848) and CBMZPN03 (grey; Lowes, M. M. J.; Cairo, M. R.; Lotter, A. P.; van der Watt, J. G. *J. Pharm. Sci.* **1987**, *76*, 744-752) generated using X'Pert HighScore Plus software.

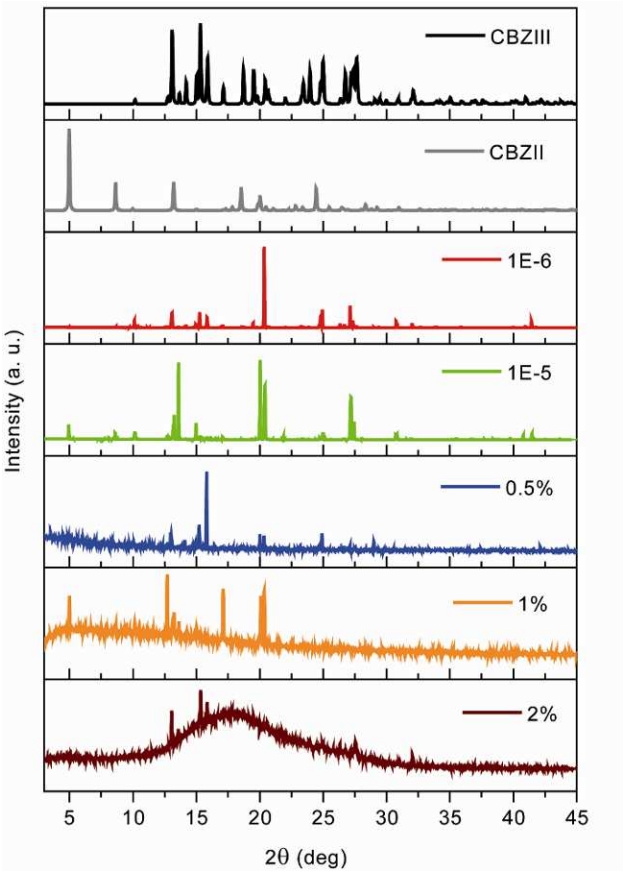


Figure S11. Comparison of the experimental XRD pattern of the crystals of CBZ obtained in the presence of organogelator **2** at different concentrations with the calculated pattern for CSD entry CBMZPN01 (black, Reboul, J. P.; Cristau, B.; Soyfer, J. C.; Astier, J. P. *Acta Crystallogr. B,* **1981,** *37*, 1844-1848) and CBMZPN03 (grey; Lowes, M. M. J.; Cairo, M. R.; Lotter, A. P.; van der Watt, J. G. *J. Pharm. Sci.* **1987**, *76*, 744-752) generated using X'Pert HighScore Plus software.

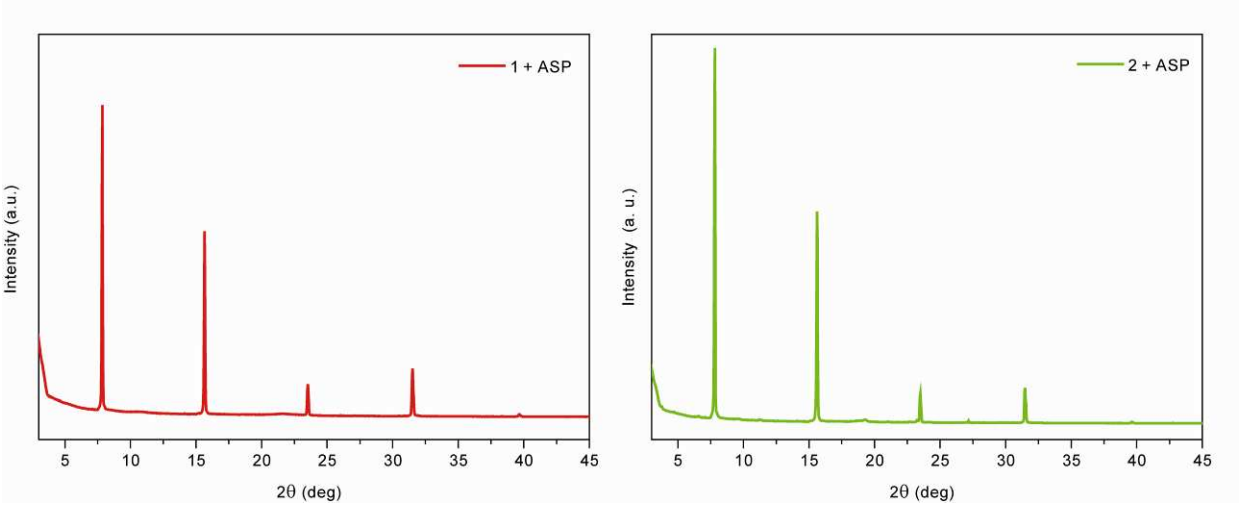


Figure S12. X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of polymorph I of aspirin obtained from the toluene gels of **1** (left), and **2** (right).

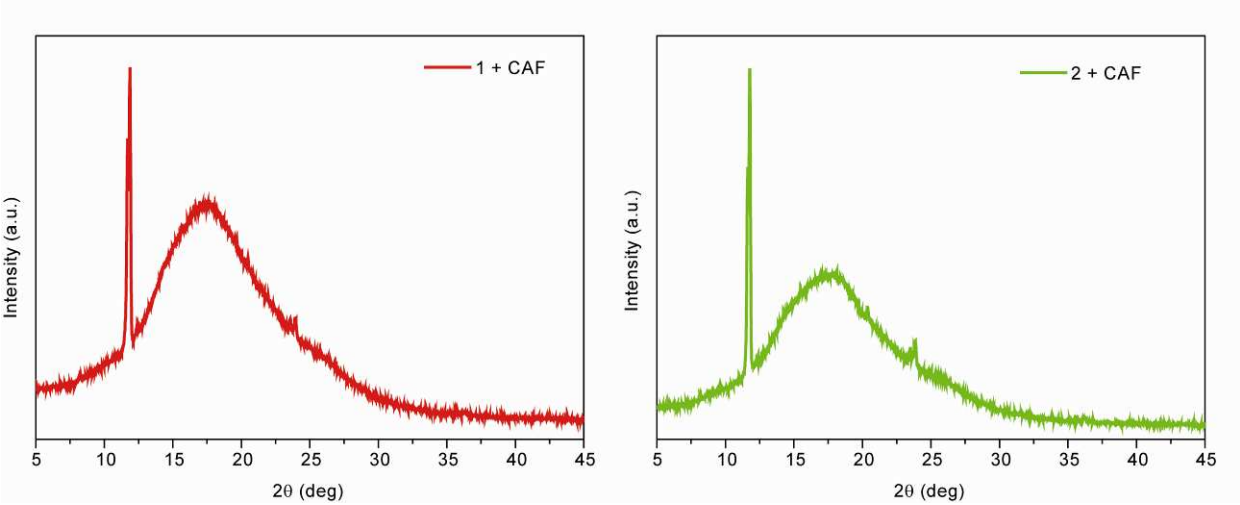


Figure S13. X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of polymorph II of caffeine obtained from the toluene gels of **1** (left), and **2** (right).

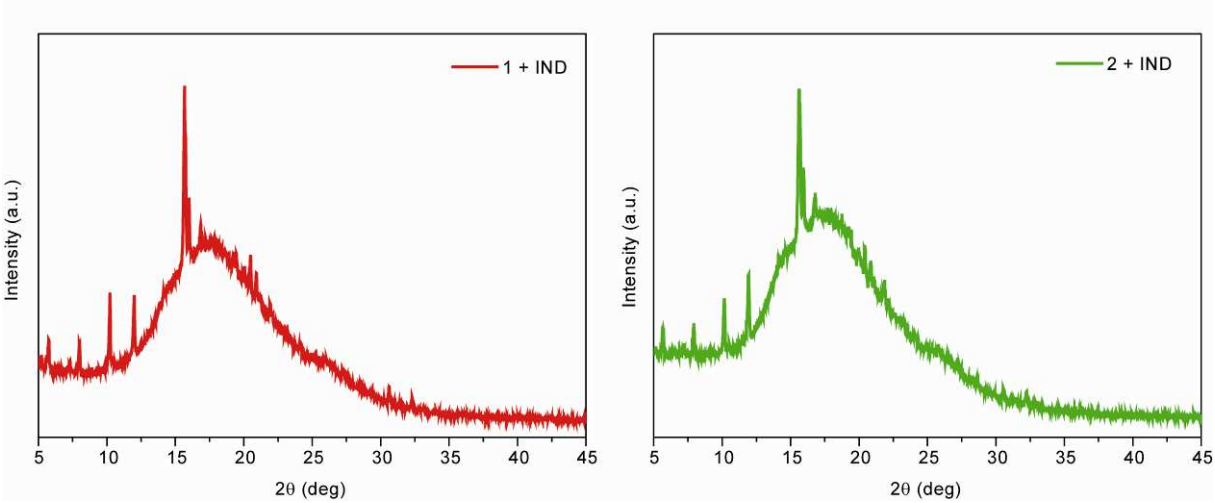


Figure S14. X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of polymorph III of indomethacin obtained from the toluenegels of **1** (left), and **2** (bottom).

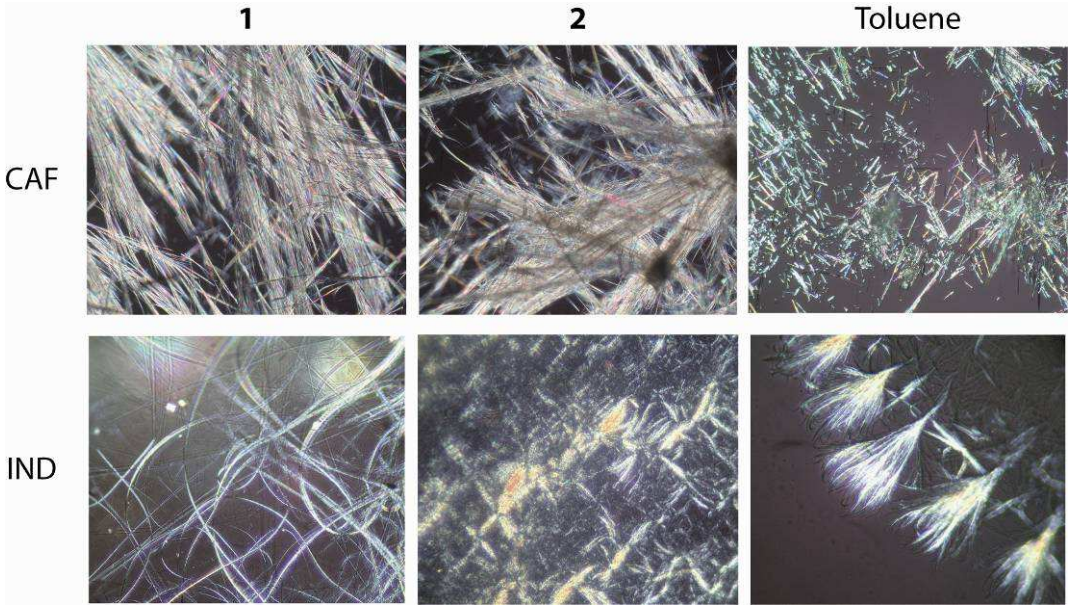


Figure S15. Optical microscopy images of CAF and IND crystals grown in organogelator **1** (left), organogelator **2** (center), and crystallized from toluene (right).

2. Tabulated X-Ray diffraction data

Table S2. X-ray diffraction data for crystals of CBZ obtained into **1** prior washing with toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
12.7139	6.95704	5.76	*III*
15.2770	5.79510	100.00	*III*
15.7024	5.63905	1.84	*III*
19.5043	4.54759	1.90	*III*
20.3512	4.36020	7.89	*III*
27.3205	3.26171	6.44	*III*
28.6162	3.11691	1.32	*III*
29.4098	3.03458	1.96	*III*
30.8555	2.89561	5.28	*III*

Table S3. X-ray diffraction data for crystals of CBZ obtained into **2** prior washing with toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
4.9398	17.87459	2.87	*II*
8.5700	10.30945	1.04	*II*
12.6472	6.99357	8.30	*III*
13.1381	6.73334	4.30	*II*
14.0856	6.28248	1.15	*III*
14.9137	5.93546	2.26	*II*
15.2284	5.81350	22.39	*III*
15.7882	5.60860	100.00	*III*
17.0220	5.20475	2.99	*II*
18.5077	4.79016	13.00	*II*
19.9891	4.43836	4.76	*II*
20.3108	4.36880	2.24	*III*
23.8503	3.72785	1.18	*III*
24.6646	3.60659	1.80	*III*
26.6226	3.34561	1.71	*III*
27.0870	3.28929	2.48	*III*
27.2534	3.26959	3.29	*III*
31.9657	2.79753	13.66	*III*

Table S4. X-ray diffraction data for crystals of CBZ obtained into the gel formed by a 9/1 mixture of **1** and **2** prior washing with toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
5.0104	17.62309	28.52	*II*
8.6588	10.20390	8.45	*II*
12.7138	6.95709	2.26	*III*
13.0598	6.77354	9.38	*III*
13.2423	6.68063	26.06	*II*
15.0267	5.89106	18.91	*III*
15.2952	5.78826	69.51	*III*
15.8681	5.58053	96.32	*III*
20.0801	4.41846	100.00	*II*
25.1659	3.53587	2.45	*III*
26.6627	3.34067	1.69	*III*
27.1349	3.28360	1.68	*III*
27.3255	3.26113	4.03	*III*
27.5187	3.23866	5.13	*III*
28.0693	3.17638	1.69	*II*
30.8778	2.89357	2.77	*III*
32.0477	2.79056	22.08	*III*
40.7932	2.21022	6.11	*III*
46.1652	1.96476	1.57	*III*

Table S5. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into organogelator **1** after washing with toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
12.7266	6.95012	1.47	*III*
13.6536	6.48028	11.67	*III*
15.3209	5.77861	100.00	*III*
15.8789	5.57677	12.11	*III*
19.4737	4.55465	1.05	*III*
27.4858	3.24247	1.39	*III*
29.4012	3.03545	4.62	*III*
30.9056	2.89103	2.82	*III*
32.0551	2.78993	1.32	*III*

Table S6. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into organogelator **2** after washing with toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
4.9964	17.67241	32.15	*II*
8.6596	10.20294	11.05	*II*
10.1598	8.69954	11.14	*II*
12.7127	6.95769	100.00	*III*
13.2414	6.68104	40.72	*II*
13.6354	6.48888	18.90	*III*
15.0353	5.88770	14.73	*III*
15.3126	5.78170	15.00	*III*
17.1212	5.17480	65.73	*II and III*
20.0910	4.41610	79.52	*II*
20.3592	4.35852	90.92	*II*
23.3979	3.79889	6.45	*II and III*
40.8937	2.20503	10.83	*III*
47.6085	1.90851	3.13	*II*

Table S7. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into the gel formed by a 9/1 mixture of organogelators **1** and **2** after washing with toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
4.9990	17.66309	35.00	*II*
8.6294	10.23858	1.03	*II*
13.0061	6.80138	1.57	*III*
13.2451	6.67918	48.83	*II*
15.0272	5.89087	24.58	*II and III*
15.3036	5.78507	49.65	*III*
15.8708	5.57959	15.51	*III*
20.0802	4.41843	100.00	*II*
23.9096	3.71874	2.83	*III*
25.1725	3.53496	2.40	*II*
26.6736	3.33932	1.51	*III*
27.1646	3.28007	1.05	*III*
27.3463	3.25870	6.40	*III*
27.5176	3.23879	7.22	*III*
30.8850	2.89291	2.40	*III*
32.0446	2.79082	4.65	*III*
34.2529	2.61578	1.45	*II and III*
36.9139	2.43309	1.59	*III*
40.8033	2.20970	4.53	*III*
46.1834	1.96403	1.21	*III*
46.5166	1.95073	1.09	*III*

Table S8. X-ray diffraction data for crystals of CBZ (1 wt%) obtained in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
10.1293	8.72566	8.85	*III*
12.6971	6.96619	2.07	*III*
15.2883	5.79084	17.10	*III*
15.8801	5.57635	10.90	*III*
17.1301	5.17215	13.68	*III*
18.7387	4.73162	1.02	*III*
20.3596	4.35842	100.00	*III*
20.5882	4.31056	1.41	*III*
23.4095	3.79704	1.28	*III*
24.7117	3.59982	0.99	*III*
24.9297	3.56883	11.88	*III*
26.6837	3.33809	1.06	*III*
27.1007	3.28766	3.45	*III*
29.9836	2.97780	1.02	*III*
30.7595	2.90443	8.61	*III*
32.0187	2.79303	1.39	*III*
39.8213	2.26190	1.09	*III*
40.8992	2.20474	2.69	*III*
40.9688	2.20115	1.86	*III*
41.4296	2.17773	7.41	*III*
46.9659	1.93311	1.38	*III*

Table S9. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-6} M solution of **1** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
4.9378	17.88193	6.14	*II*
8.6170	10.25338	5.14	*II*
10.1277	8.72704	1.73	*III*
12.6604	6.98634	0.97	*III*
13.0960	6.75488	100.00	*II and III*
13.2224	6.69061	4.35	*II*
13.6157	6.49823	7.42	*III*
14.9954	5.90330	9.86	*II and III*
15.2716	5.79715	61.22	*III*
15.8432	5.58925	11.65	*III*
17.0732	5.18925	1.09	*II and III*
18.7154	4.73747	1.77	*III*
19.4901	4.55086	2.31	*III*
19.9881	4.43860	13.47	*II*
20.3509	4.36027	12.46	*III*
21.9636	4.04362	3.04	*III*
23.4028	3.79811	1.97	*II and III*
23.9007	3.72011	1.50	*III*
24.7212	3.59846	1.94	*III*
24.9048	3.57235	18.46	*III*
26.3485	3.37979	16.05	*II and III*
27.1451	3.28239	7.44	*III*
27.2513	3.26983	1.29	*III*
27.5027	3.24051	4.38	*III*
29.8809	2.98780	1.44	*III*
30.8565	2.89552	4.00	*II and III*
32.0096	2.79380	1.33	*III*
32.7069	2.73581	1.46	*III*
39.9662	2.25403	1.93	*II and III*
40.4316	2.22915	1.52	*III*
46.4350	1.95397	2.04	*III*

Table S10. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-5} M solution of **1** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
10.1166	8.74385	5.77	*III*
12.6802	6.98124	9.34	*III*
13.0213	6.79913	16.52	*III*
13.5964	6.51278	13.11	*III*
14.1385	6.26426	4.73	*III*
14.9447	5.92810	3.86	*III*
15.2684	5.80316	100.00	*III*
15.8452	5.59319	60.63	*III*
17.0701	5.19448	4.46	*III*
18.6514	4.75751	10.84	*III*
19.4693	4.55945	39.84	*III*
19.7484	4.49564	1.83	*III*
20.3530	4.36344	39.78	*III*
20.5658	4.31878	3.53	*III*
21.9376	4.05171	1.69	*III*
23.2217	3.83049	1.03	*III*
23.3612	3.80793	8.57	*III*
23.5690	3.77482	1.22	*III*
23.8816	3.72611	12.90	*III*
24.7200	3.60161	14.44	*III*
24.8952	3.57666	9.78	*III*
26.6379	3.34650	1.85	*III*
27.1313	3.28674	10.04	*III*
27.2880	3.26822	5.43	*III*
27.5701	3.23542	6.74	*III*
29.3791	3.04019	1.09	*III*
30.7498	2.90533	2.17	*III*
30.8632	2.89731	2.82	*III*
32.0262	2.79470	14.13	*III*
34.9525	2.56713	3.35	*III*
41.4247	2.17978	1.50	*III*
46.9446	1.93394	1.22	*III*

Table S11. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-3} M solution of **1** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
10.1193	8.73424	8.62	*III*
12.6788	6.97621	6.08	*III*
13.0420	6.78274	75.90	*III*
13.6019	6.50480	59.47	*III*
14.1414	6.25780	15.83	*III*
14.9624	5.91625	25.55	*III*
15.2601	5.80150	32.93	*III*
15.8180	5.59810	7.85	*III*
17.0226	5.20457	2.84	*III*
18.6458	4.75500	13.20	*III*
19.4466	4.56094	16.30	*III*
20.3515	4.36014	100.00	*III*
20.5586	4.31670	5.36	*III*
23.3688	3.80356	9.13	*III*
23.8542	3.72725	6.26	*III*
24.7034	3.60101	12.89	*III*
24.8952	3.57370	12.25	*III*
26.2942	3.38664	10.46	*III*
26.6488	3.34238	15.32	*III*
27.1153	3.28593	5.46	*III*
27.2920	3.26505	31.30	*III*
27.4947	3.24144	44.47	*III*
29.3596	3.03965	2.83	*III*
30.7513	2.90518	10.66	*III*
32.0127	2.79353	2.68	*III*
34.9347	2.56628	3.81	*III*
41.4212	2.17815	9.77	*III*
46.0578	1.96909	1.40	*III*
46.4287	1.95422	4.97	*III*

Table S12. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 5 x 10^{-3} M (1 wt%) solution of **1** in toluene.

$2\theta_{exp}$ (°)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
12.7139	6.95704	4.42	*III*
15.2770	5.79510	100.00	*III*
15.7024	5.63905	1.25	*III*
20.3512	4.36020	7.86	*III*
27.3205	3.26171	6.29	*III*
29.4098	3.03458	1.42	*III*
30.8555	2.89561	5.30	*III*

Table S13. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-2} M (2 wt%) solution of **1** in toluene.

$2\theta_{exp}$ (°)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
12.9050	6.85442	100.00	*III*
14.0938	6.27885	6.27	*III*
14.8461	5.96233	18.36	*III*
15.1559	5.84115	25.06	*III*
15.7183	5.63339	13.46	*III*
16.9654	5.22197	9.97	*III*
18.4848	4.79603	11.40	*III*
19.3296	4.58828	15.51	*III*
20.2389	4.38414	31.29	*III*
23.2154	3.82835	6.92	*III*
23.7486	3.74358	17.11	*III*
24.7206	3.59855	72.34	*III*
24.8107	3.58568	82.03	*III*
26.1611	3.40357	17.78	*III*
26.4956	3.36136	26.10	*III*
27.2280	3.27259	35.43	*III*
27.4261	3.24940	23.54	*III*
29.7539	3.00026	2.50	*III*
30.6691	2.91278	3.33	*III*
31.8906	2.80395	3.70	*III*
32.5537	2.74833	2.85	*III*
33.8182	2.64840	8.13	*III*
39.7607	2.26520	4.69	*III*
41.9825	2.15032	3.38	*III*

Table S14. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-6} M solution of **2** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
8.6622	10.19992	1.23	*II*
10.1229	8.73116	9.66	*III*
12.6754	6.97807	1.21	*III*
13.0865	6.75976	18.67	*III*
14.1792	6.24120	2.25	*III*
14.9716	5.91260	4.06	*II and III*
15.2669	5.79892	18.67	*III*
15.8537	5.58557	13.05	*III*
17.0887	5.18458	2.15	*II and III*
18.6146	4.76289	1.04	*III*
19.4592	4.55801	5.16	*III*
20.3570	4.35897	100.00	*III*
20.5875	4.31069	1.10	*III*
23.2397	3.82439	1.44	*III*
24.8002	3.58718	14.48	*III*
24.9396	3.56744	22.40	*III*
26.3266	3.38255	3.28	*III*
26.6903	3.33728	2.63	*III*
27.1364	3.28342	27.35	*III*
27.3374	3.25974	9.11	*III*
27.4942	3.24150	1.61	*III*
27.6471	3.22392	1.23	*III*
30.7583	2.90454	8.64	*III*
32.0300	2.79206	4.19	*III*
40.9234	2.20349	1.71	*III*
41.4308	2.17767	8.06	*III*

Table S15. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-5} M solution of **2** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
4.9468	17.84946	18.42	*II*
8.6123	10.25896	7.87	*II*
10.1834	8.67945	8.10	*III*
12.6942	6.96779	3.86	*III*
12.8108	6.90465	1.78	*III*
13.2558	6.67382	29.74	*II*
13.6024	6.50457	98.61	*III*
14.1660	6.24699	1.03	*III*
14.9785	5.90990	20.63	*II and III*
15.3262	5.77662	4.92	*III*
15.8503	5.58676	2.49	*III*
17.1070	5.17907	2.66	*II and III*
20.0320	4.42896	100.00	*II*
20.3970	4.35051	66.68	*III*
21.8649	4.06166	3.83	*II*
24.7200	3.59863	2.29	*III*
24.9993	3.55905	9.19	*III*
25.1272	3.54123	3.13	*II*
27.2087	3.27486	49.78	*III*
27.4421	3.24754	22.48	*III*
28.0557	3.17789	1.36	*II and III*
30.7193	2.90814	5.20	*II*
30.8099	2.89979	6.65	*II and III*
31.9530	2.79862	1.12	*III*
35.4864	2.52763	1.15	*II*
39.7728	2.26454	1.45	*II and III*
40.7629	2.21180	4.56	*II and III*
41.4595	2.17623	7.25	*II and III*
45.0528	2.01065	1.15	*III*
46.1419	1.96570	1.24	*III*

Table S16. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-3} M (0.5 wt%) solution of **2** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
4.9369	17.88516	5.38	*II*
12.6695	6.98135	3.68	*III*
12.9999	6.80464	15.25	*III*
14.0924	6.27947	9.73	*III*
14.9494	5.92135	7.99	*II and III*
15.2267	5.81415	29.51	*III*
15.8119	5.60026	100.00	*III*
17.0061	5.20957	2.87	*III*
18.6368	4.75725	5.78	*II and III*
19.4106	4.56932	1.77	*III*
20.0207	4.43143	17.09	*II*
20.3468	4.36115	16.49	*III*
21.9052	4.05428	1.03	*II and III*
23.3504	3.80651	2.23	*II and III*
23.8417	3.72918	1.89	*III*
24.6898	3.60297	3.03	*III*
24.9037	3.57250	19.19	*III*
26.2764	3.38889	1.79	*III*
26.6394	3.34354	3.52	*II and III*
27.1027	3.28742	4.30	*III*
27.5594	3.23397	3.24	*III*
29.0025	3.07626	10.14	*III*
29.8362	2.99217	1.27	*III*
37.4529	2.39931	1.95	*II and III*
37.7031	2.38396	1.00	*III*
40.7514	2.21239	1.79	*II and III*
42.1244	2.14340	4.68	*II and III*

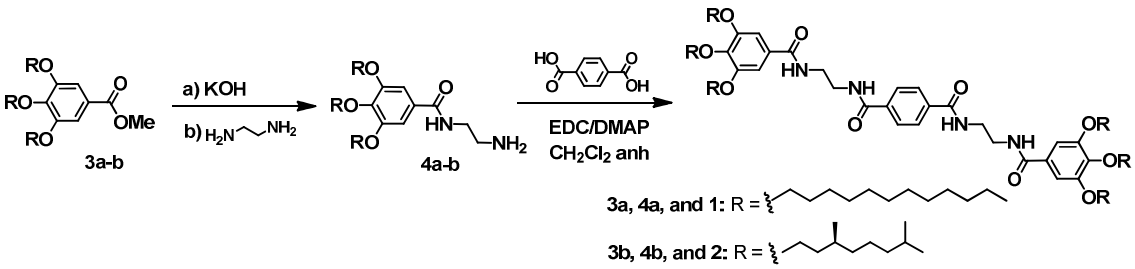
Table S17. X-ray diffraction data for crystals of CBZ (1 wt%) obtained into a 1 x 10^{-2} M (2 wt%) solution of **2** in toluene.

$2\theta_{exp}$ (º)	d_{exp} (Å)	Rel. Int. (%)	Polymorph
13.0541	6.77647	95.39	*III*
13.5850	6.51285	27.08	*III*
14.1324	6.26175	4.98	*III*
15.3530	5.76657	100.00	*III*
15.8579	5.58409	57.56	*III*
17.1792	5.15746	6.43	*III*
18.7267	4.73462	14.02	*III*
19.4902	4.55084	11.14	*III*
20.6008	4.30795	18.58	*III*
23.8734	3.72429	15.99	*III*
24.9753	3.56242	12.43	*III*
26.3065	3.38509	22.30	*III*
26.7166	3.33405	7.31	*III*
27.1775	3.27855	13.26	*III*
27.4965	3.24123	31.17	*III*
27.6091	3.22827	28.20	*III*
29.4469	3.03083	4.35	*III*
30.9107	2.89057	3.97	*III*
32.0414	2.79109	25.04	*III*
34.9688	2.56385	3.17	*III*
40.9129	2.20403	2.90	*III*

3. Experimental section

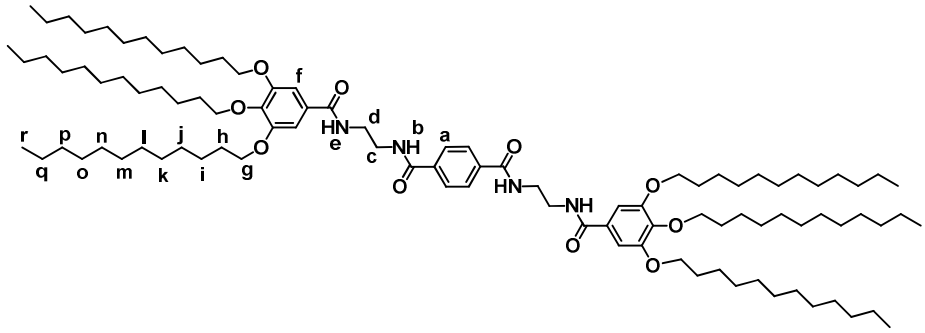
General. All solvents were dried according to standard procedures. Reagents were used as purchased. All air-sensitive reactions were carried out under argon atmosphere. Flash chromatography was performed using silica gel (Merck, Kieselgel 60, 230-240 mesh or Scharlau 60, 230-240 mesh). Analytical thin layer chromatography (TLC) was performed using aluminium-coated Merck Kieselgel 60 F254 plates. NMR spectra were recorded on a Bruker Avance 300 (^{1}H: 300 MHz; ^{13}C: 75 MHz) spectrometer at 298 K using partially deuterated solvents as internal standards. Coupling constants (J) are denoted in Hz and chemical shifts (δ) in ppm. Multiplicities are denoted as follows: s = singlet, d = doublet, t = triplet, m = multiplet, br = broad. FT-IR spectra were recorded on a Bruker Tensor 27 (ATR device) spectrometer. UV-Vis spectra were recorded on a Varian Cary 50 spectrophotometer. High resolution mass spectra (HRMS) were recorded on a FTMS Bruker APEX Q IV spectrometer. Circular dichroism (CD) measurements were performed on a Jasco-810 dichrograph equipped with a Peltier thermoelectric temperature controller. The spectra were recorded in the continuous mode between 350 and 200 nm, with a wavelength increment of 1 nm, a response time of 4 s, and a bandwidth of 1 nm. A 1 cm path length quartz cuvette (Hellma) was used. Spectra from three scans were averaged. Thermal experiments were performed at constant heating rates of 1 K/min by following the ellipticity at 222 nm from 10 to 90 ℃ in 1 cm path length q uartz cuvette (Hellma) with a total sample concentration of 1×10^{-5} M in methylcyclohexane. Scanning electron microscopy images were obtained from on a JEOL JSM 6335F microscope working at 10kV. Atomic force microscopy was performed on a SPM Nanoscope IIIa multimode microscope working on tapping mode with a RTESPA tip (Veeco) at a working frequency of ~235 kHz. All the crystallization experiments were carried out per duplicate. X-ray diffraction was performed in a Panalytical X'Pert PRO diffractometer with Cu tube and primary beam monochromator (lambda K□=1.5406 Å) operated at 45 kV, 40 mA, programmable divergence slit working in fixed mode, and fast linear detector (X'Celerator) working in scanning mode. Samples were deposited on "zero background" silicon sample holders and measured in reflection geometry. To prevent solvent evaporation in gel samples, the sample holders were enclosed in a cell with a x-ray window covered by a kapton foil.

4. Synthetic details and characterization



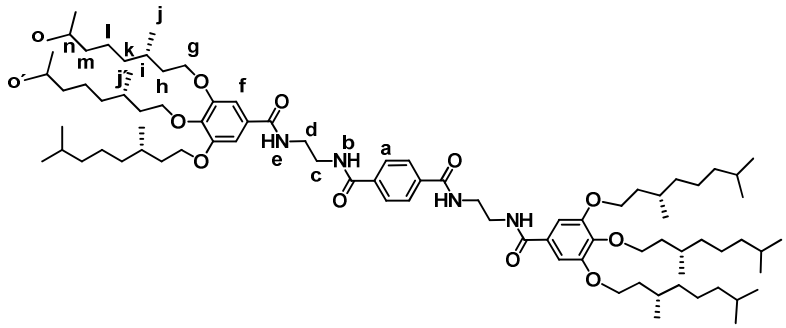
Terephthalic acid was purchased from a commercial source. Monoamides **4** were prepared according to previously reported synthetic procedures (see: Dawn, A.; Fujita, N.; Haraguchi, S.; Sada, K.; Shinkai S. Chem. Commun. **2009**, 2100, and Ghosh, S.; Li, X.; Stepanenko, V.; Würthner, F. Chem. Eur. J. **2008**, 14, 11343, respectively) and showed identical spectroscopic properties to those reported therein.

N^1,N^4-bis(2-(3,4,5-tridecyloxybenzamido)ethyl)terephthalamide (1)



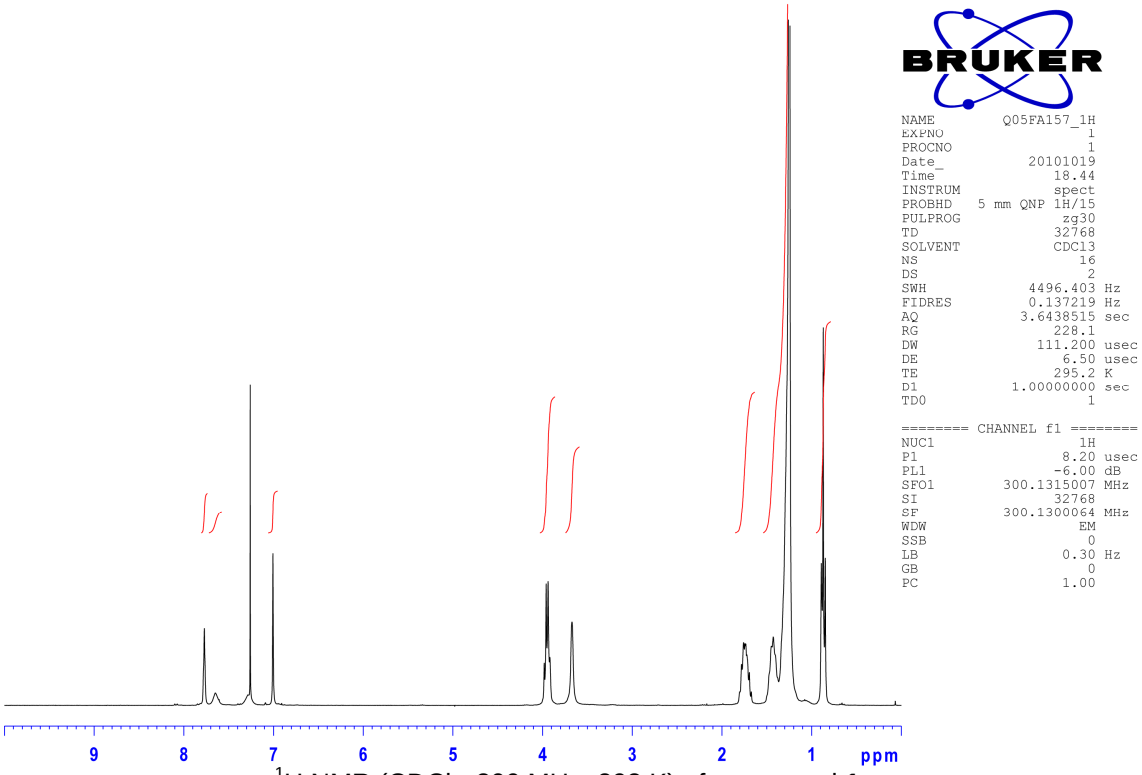
A solution of 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride (0.07 g, 0.35 mmol) and 4-dimethylaminopyridine (0.04 g, 0.35 mmol) in dry methylene chloride (2 mL) was added to a previously prepared solution of terephthalic acid (0.03 g, 0.16 mmol) in dry THF (2 mL). The resulting mixture was cooled to 0 ºC and stirred for 15 minutes under argon atmosphere. After that, **4a** (0.25 g, 0.35 mmol) was added portionwise. The reaction mixture was stirred at room temperature for 16 hours. The organic layer was washed with HCl 1M, $NaHCO_3$ 1M, and water, and dried over $MgSO_4$. After evaporation of the solvent, the residue was purified by column chromatography (silica gel, chloroform: methanol 95:5) affording compound **1** as a white solid (0.15 g, 58%). ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.77 (4H, H_a, br), 7.65 (2H, H_b, br) 7.28 (2H, H_e, br), 7.00 (4H, H_f, s), 3.95 (12H, H_g, m), 3.67 (8H, H_{c+d}, br), 1.74 (12H, H_h, m), 1.43 (12H, H_i, m), 1.25 (96H, H_{j-q}, br), 0.87 (18H, H_r, t, J=6.8 Hz) ;^{13}C NMR ($CDCl_3$, 75Mz) δ 169.0, 167.8, 153.3, 141.6, 136.9, 128. 7, 127.5, 105.9, 73.7, 69.5, 41.5, 41.0, 32.1, 30.5, 29.8, 29.5, 26.3, 22.8, 14. 2; FTIR (neat) 712, 792, 821, 1168, 1232, 1286, 1345, 1386, 1427, 1468, 1546, 1581, 1633, 2853, 2922, 3274 cm^{-1}; HRMS (ESI-FT): calcd. for $C_{98}H_{170}N_4O_{10}$ $[M]^+$, 1563.38165; found, 1563.38256.

N^1,N^4-bis(2-(3,4,5-tri-((*S*)-3',7'-dimethyloctyl)oxybenzamido)ethyl)terephthalamide (2)

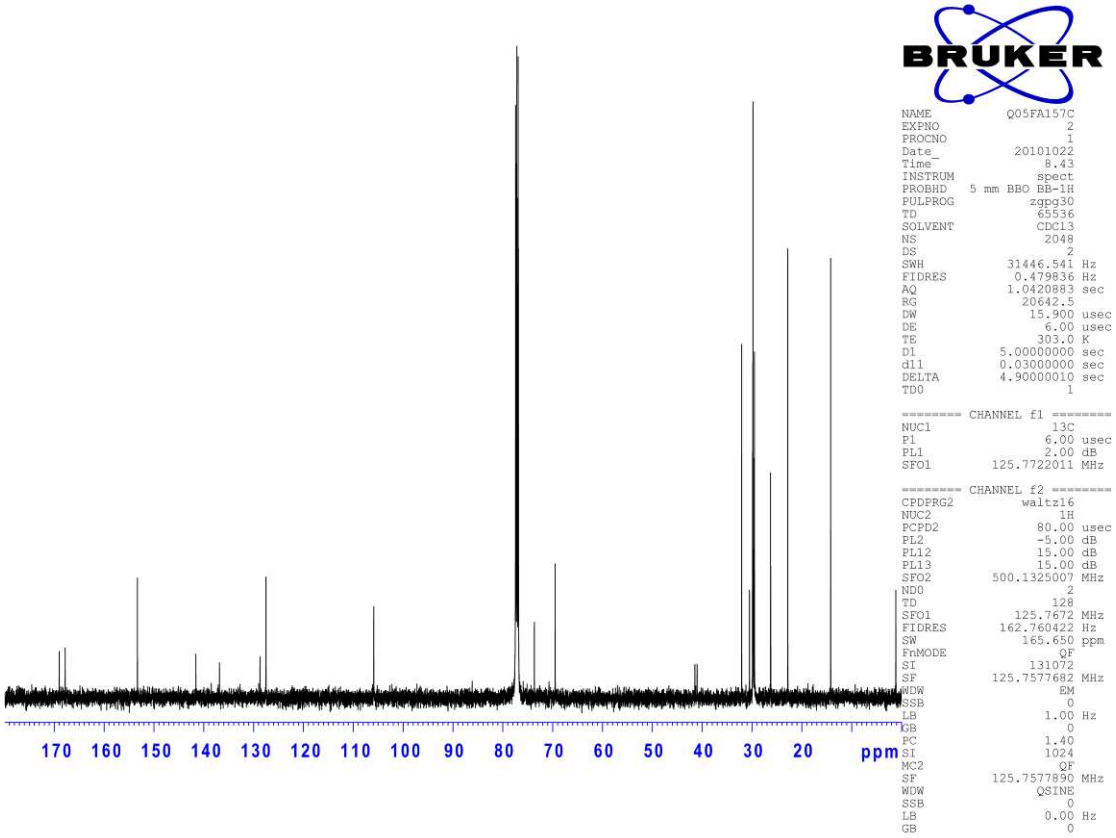


A solution of 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride (0.08 g, 0.40 mmol) and 4-dimethylaminopyridine (0.05 g, 0.40 mmol) in dry methylene chloride (2 mL) was added to a previously prepared solution of terephthalic acid (0.03 g, 0.18 mmol) in dry THF (2 mL). The resulting mixture was cooled to 0 ºC and stirred for 15 minutes under argon atmosphere. After that, **4b** (0.26 g, 0.40 mmol) was added portionwise. The reaction mixture was stirred at room temperature for 16 hours. The organic layer was washed with HCl 1M, $NaHCO_3$ 1M, and water, and dried over $MgSO_4$. After evaporation of the solvent, the residue was purified by column chromatography (silica gel, chloroform: methanol 95:5) affording compound **2** as a white solid (0.10 g, 42%). ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.74 (6H, H_{a+b}, br), 7.44 (2H, H_e, br) 7.04 (4H, H_f, s), 3.98 (12H, H_g, m), 3.66 (8H, H_{c+d}, br), 1.82 (6H, H_i, m), 1.67 (6H, H_n, br), 1.50 (12H, H_h, m), 1.38-1.04 (36H, H_{k-m}, br), 0.90 (6H, $H_{j'}$, d, J=6.5 Hz), 0.89 (12H, H_j, d, J=6.5 Hz), 0.85 (12H, $H_{o'}$, d, J=6.6 Hz), 0.84 (24H, H_o, d, J=6.6 Hz);^{13}C NMR ($CDCl_3$, 75Mz) δ 168.8, 168.0, 153.2, 141.3, 136.8, 128. 8, 127.5, 105.7, 71.9, 67.6, 41.1, 41.0, 39.5, 39.4, 37.7, 37.5, 36.5, 29.9, 29.8, 28.1, 24.9, 22.8, 22.7, 19.7, 19.6; FTIR (neat) 704, 859, 1118, 1232, 1296, 1342, 1380, 1431, 1466, 1498, 1543, 1581, 1635, 2925, 2953, 3288 cm^{-1}; HRMS (ESI-FT) calcd. for $C_{86}H_{146}N_4NaO_{10}$ $[M+Na]^+$, 1418.09312; found, 1419.09799.

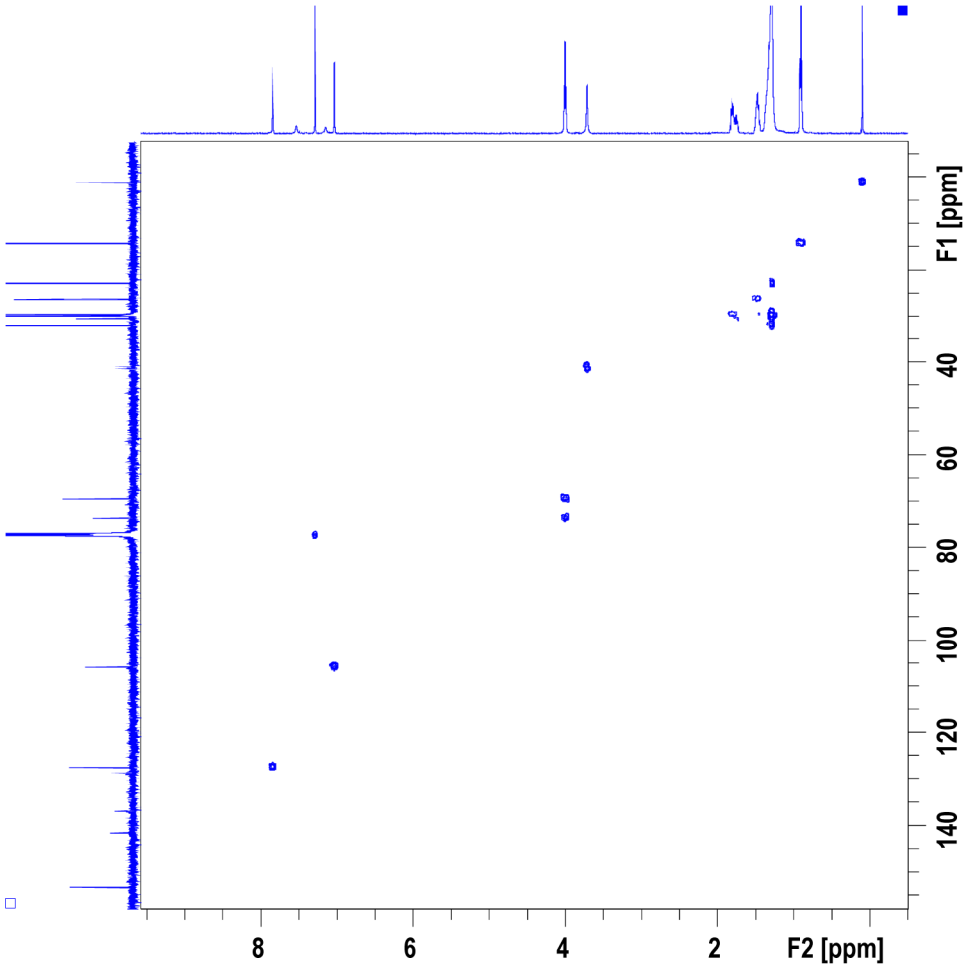
5. Collection of spectra



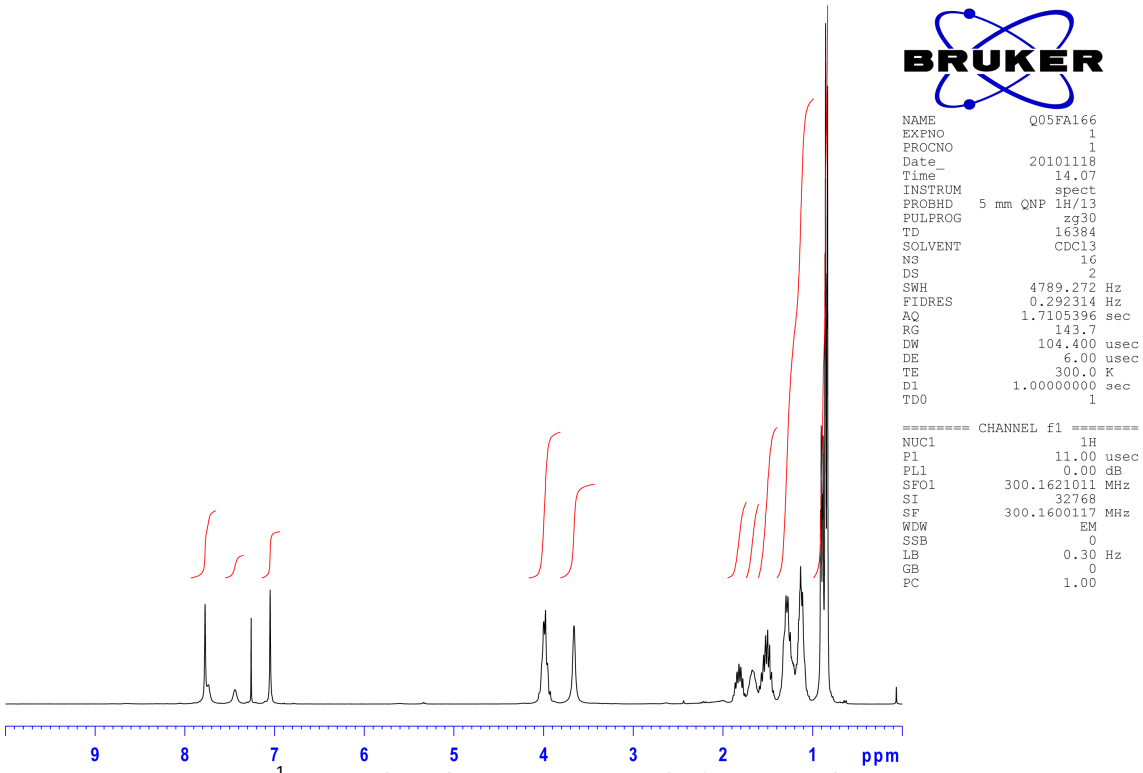
^{1}H NMR ($CDCl_3$, 300 MHz, 298 K) of compound **1**.



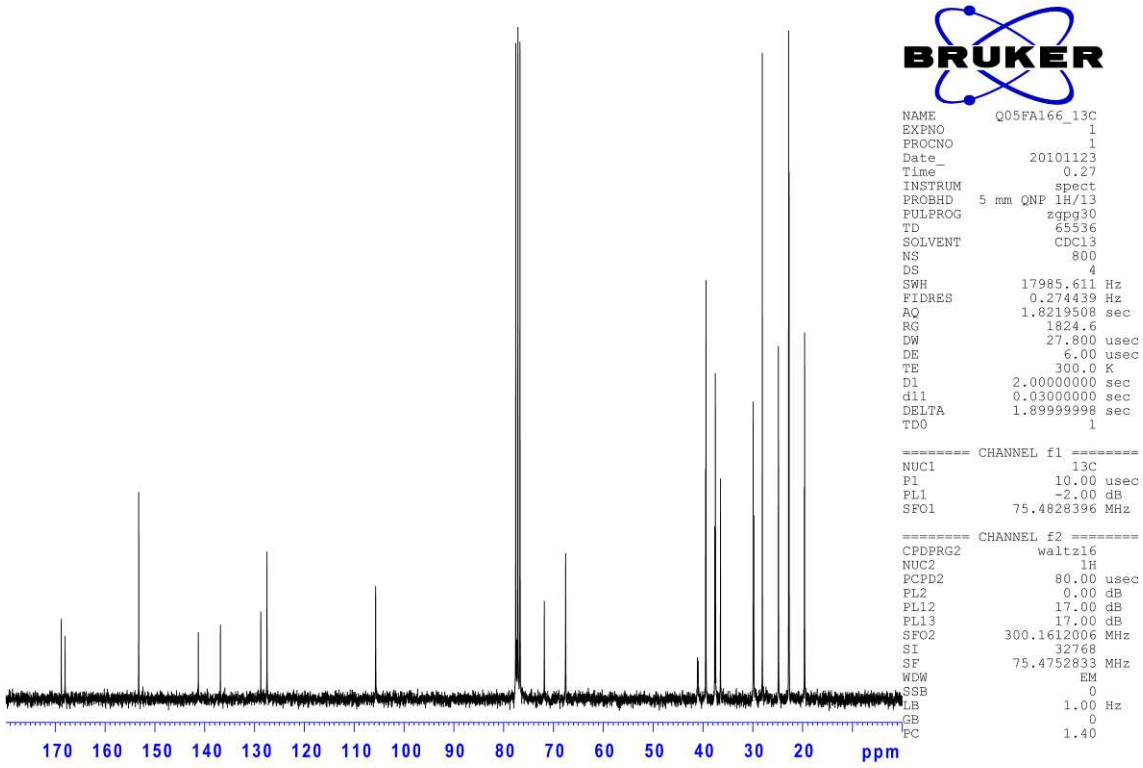
^{13}C NMR ($CDCl_3$, 75 MHz, 298 K) of compound **1**.



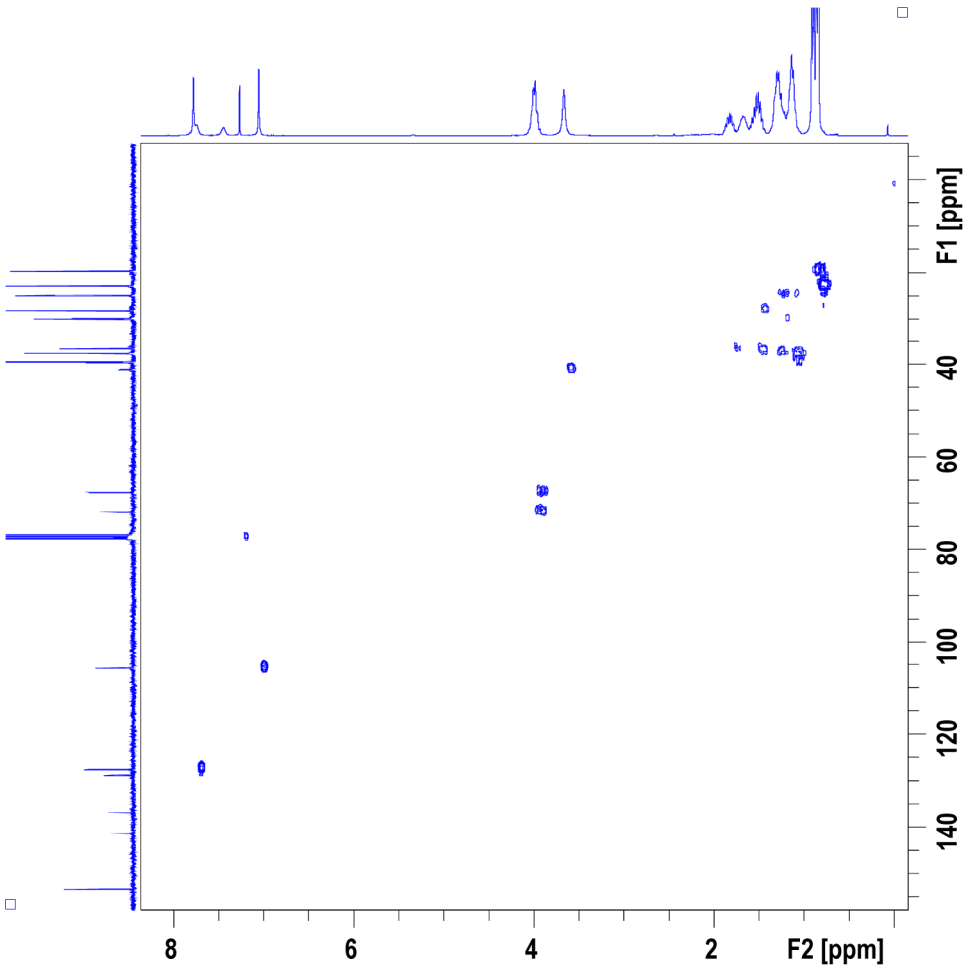
^{1}H, ^{13}C-HMQC spectrum ($CDCl_3$, 298 K) of compound **1**.



^{1}H NMR ($CDCl_3$, 300 MHz, 298 K) of compound **2**.



^{13}C NMR ($CDCl_3$, 75 MHz, 298 K) of compound **2**.



^{1}H, ^{13}C-HMQC spectrum ($CDCl_3$, 298 K) of compound **2**.