Electronic Supplementary Information (ESI)

Self-assembled sonogels formed from 1,4-naphthalenedicarbonyldinicotinic acid hydrazide

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1. Experimental Section

1.1 Synthesis

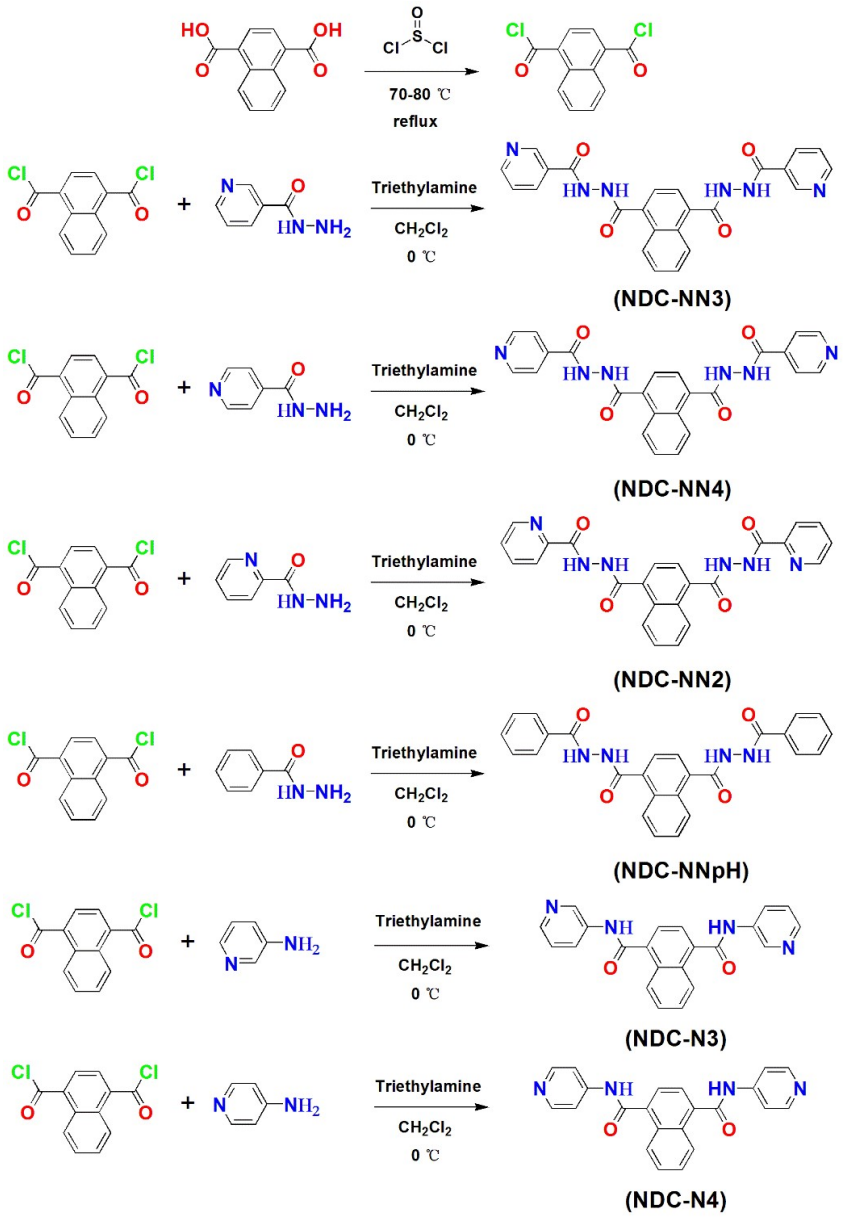
The compound 1,4-naphthalenedicarbonyldinicotinic acid hydrazide (NDC-NN3) was prepared according to the literature method.[1] The compounds of 1,4-naphthalenedicarbonyldiisonicotinic acid hydrazide (NDC-NN4), 1,4-naphthalenedicarbonyldipyridine-2-carboxylic acid hydrazide (NDC-NN2), 1,4-naphthalenedicarbonyldibenzoic acid hydrazide (NDC-NNpH), N^1,N^4-di(pyridin-3-yl)-naphthalene-1,4-dicarboxamide (NDC-N3) and N^1,N^4-di(pyridin-4-yl)-naphthalene-1,4-dicarboxamide (NDC-N4) were prepared similar to NDC-NN3. In a typical experiment, 1,4-Naphthalenedicarbonyl dichloride was prepared from 1,4-Naphthalenedicarboxylic acid by treatment with $SOCl_2$ using a catalytic amount of DMF. Subsequently, a DCM (100mL) solution of naphthalene-1,4-dicarbonyl dichloride (2.53 g, 10.0 mmol) was then added dropwise to a DCM (100mL) solution of nicotinic acid hydrazide (2.88 g, 21.0 mmol) and triethylamine (3.0 mL, 29.6 mmol) with continuous stirring in an ice bath. After stirring for 24h, the pale yellow precipitate was filtered and washed with DCM for four times. The product was recrystallized from a mixed solvent of H_2O (300 mL) and ethanol (100 mL), and then dried under vacuum to afford 3.7 g of NDC-NN3 (yield 70%). The compounds prepared in this work were characterized by FT-IR, ^{1}H NMR, ^{13}C NMR and LC-MS.

1,4-naphthalenedicarbonyldinicotinic acid hydrazide (NDC-NN3)

^{1}H NMR (400MHz, DMSO-*d*6, δ_H): 10.92 (2H, s), 10.71 (2H, s), 9.14 (2H, *J* = 1.6 Hz, d), 8.81 (1 H, *J* = 4.8, 1.7 Hz, dd), 8.48 (2H, *J* = 6.5, 3.3 Hz, dd), 8.39 – 8.31 (2H, m), 7.78 (2H, s), 7.72 (2H, *J* = 6.5, 3.3 Hz, dd), 7.62 (2H, *J* = 7.9, 4.8 Hz, dd). ^{13}C NMR (100MHz, DMSO-*d6*, δ_C): 168.15, 165.04, 153.15, 148.97, 135.83, 135.34, 130.49, 128.53, 127.89, 126.20, 125.04, 124.30. IR (KBr): 3234, 1658, 1594, 1515, 1474, 1421, 1293, 1261, 1172, 1140, 1028, 907, 825, 772, 705, 485 cm^{-1}. LC-MS $[M+H]^+$ calculated for $C_{17}H_{13}N_5O_2$: m/z 454.1389; found: 455.1476.

1,4-naphthalenedicarbonyldiisonicotinic acid hydrazide (NDC-NN4)

^{1}H NMR (400MHz, DMSO-*d*6, δ_H): 11.02 (2H, s), 10.74 (2H, s), 8.88 – 8.81 (4H, m), 8.47 (2H, *J* = 6.5, 3.3 Hz, dd), 7.90 (4H, *J* = 4.4, 1.6 Hz, dd), 7.78 (2H, s), 7.73 (2H, *J* = 6.5, 3.3 Hz, dd). ^{13}C NMR (100MHz, DMSO-*d6*, δ_C): 168.07, 164.91, 151.04, 139.84, 135.30, 130.49, 127.93, 126.19, 125.07, 121.89. IR (KBr): 3425, 3177, 1661, 1603, 1553, 1515, 1410, 1293, 1259, 1141, 1064, 846, 778, 675, 469 cm^{-1}. LC-MS $[M+H]^+$ calculated for $C_{17}H_{13}N_5O_2$: m/z 454.1389; found: 455.1475.



Scheme S1. Synthetic routes for gelators (NDC-NN3, NDC-NN4, NDC-NN2, NDC-NNpH, NDC-N3 and NDC-N4) in this work

1,4-naphthalenedicarbonyldipyridine-2-carboxylic acid hydrazide (NDC-NN2)

^{1}H NMR (400MHz, DMSO-*d*6, δ_H): 10.85 (2H, s), 10.66 (2H, s), 8.75 (2H, *J* = 4.6 Hz, d), 8.49 (2H, *J* = 6.5, 3.3 Hz, dd), 8.15 (2H, *J* = 7.7 Hz, d), 8.09 (1 H, *J* = 7.6, 1.7 Hz, td), 7.75 (2H, s), 7.73 – 7.65 (4H, m). ^{13}C NMR (100MHz, DMSO-*d6*, δ_C): 167.79, 163.85, 149.67, 149.23, 138.41, 135.46, 130.53, 127.66, 127.60, 126.34, 124.93, 123.98. IR (KBr): 3381, 3260, 1677, 1651, 1590, 1571, 1490, 1431, 1283, 1242, 1140, 1040, 858, 816, 747, 620 cm^{-1}. $[M+H]^+$ calculated for $C_{17}H_{13}N_5O_2$: m/z 454.1389; found: 455.1464.

1,4-naphthalenedicarbonyldibenzoic acid hydrazide (NDC-NNpH)

^{1}H NMR (400MHz, DMSO-*d*6, δ_H): 10.73 (2H, s), 10.64 (2H, s), 8.51 (2H, *J* = 6.5, 3.3 Hz, dd), 8.07 – 7.95 (4H, m), 7.78 (2H, s), 7.73 (2H, *J* = 6.5, 3.3 Hz, dd), 7.68 – 7.61 (2H, m), 7.57 (4H, *J* = 11.4, 4.4 Hz, dd). ^{13}C NMR (100MHz, DMSO-*d6*, δ_C): 168.20, 166.33, 135.50, 132.86, 132.49, 130.53, 129.07, 128.01, 127.77, 126.28, 124.98. IR (KBr): 3424, 3179, 1655, 1601, 1575, 1545, 1473, 1287, 1258, 1076, 710, 477 cm^{-1}. $[M+Na]^+$ calculated for $C_{26}H_{20}N_4O_4$: m/z 452.1485; found: 475.1378.

N^1,N^4-di(pyridin-3-yl)-naphthalene-1,4-dicarboxamide (NDC-N3)

^{1}H NMR (400MHz, DMSO-*d*6, δ_H): 10.88 (2H, s), 8.97 (2H, *J* = 2.1 Hz, d), 8.37 (2H, *J* = 4.7, 1.2 Hz, dd), 8.32 – 8.21 (4H, m), 7.90 (2H, s), 7.70 (2H, *J* = 6.5, 3.3 Hz, dd), 7.46 (2H, *J* = 8.3, 4.7 Hz, dd). ^{13}C NMR (100MHz, DMSO-*d6*, δ_C): 167.72, 145.31, 142.03, 136.74, 136.27, 130.22, 127.45, 126.05, 125.09, 124.22. IR (KBr): 3424, 3238, 1674, 1652, 1600, 1546, 1479, 1426, 1331, 1310, 1271, 880, 836, 793, 770, 705, 627, 473 cm^{-1}. LC-MS $[M+H]^+$ calculated for $C_{17}H_{13}N_5O_2$: m/z 368.1273; found: 369.1357.

N^1,N^4-di(pyridin-4-yl)-naphthalene-1,4-dicarboxamide (NDC-N4)

^{1}H NMR (400MHz, DMSO-*d*6, δ_H):11.08 (2H, s), 8.54 (4H, *J* = 4.8, 1.5 Hz, dd), 8.23 (1H, *J* = 6.5, 3.3 Hz, dd), 7.90 (1H, s), 7.80 (2H, dd, *J* = 4.9, 1.4 Hz, dd), 7.71 (1H, *J* = 6.5, 3.3 Hz, dd). ^{13}C NMR (100MHz, DMSO-*d6*, δ_C): 168.17, 150.95, 146.17, 136.64, 130.08, 128.17, 125.92, 125.14, 114.28. IR (KBr): 3291, 3225, 3153，3062, 2967, 2926，1685, 1594, 1514, 1418, 1331, 1301, 1252, 1208, 1181, 1146, 1029, 996, 886, 829,768 cm^{-1}. LC-MS $[M+H]^+$ calculated for $C_{22}H_{16}N_4O_2$: m/z 368.1273; found: 369.1346.

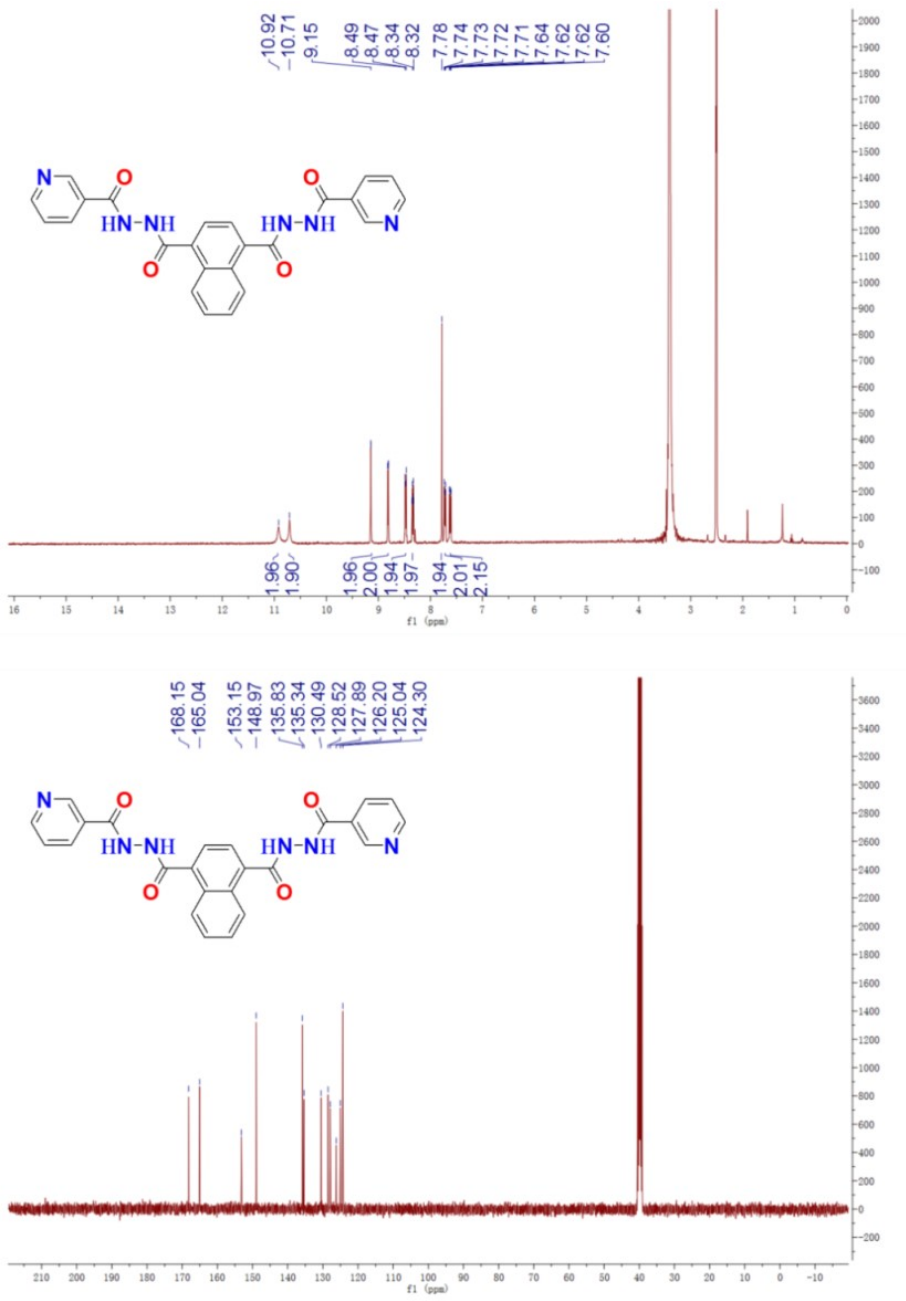


Fig. S1. ^{1}H NMR and ^{13}C NMR spectra of NDC-NN3 in DMSO-*d*6

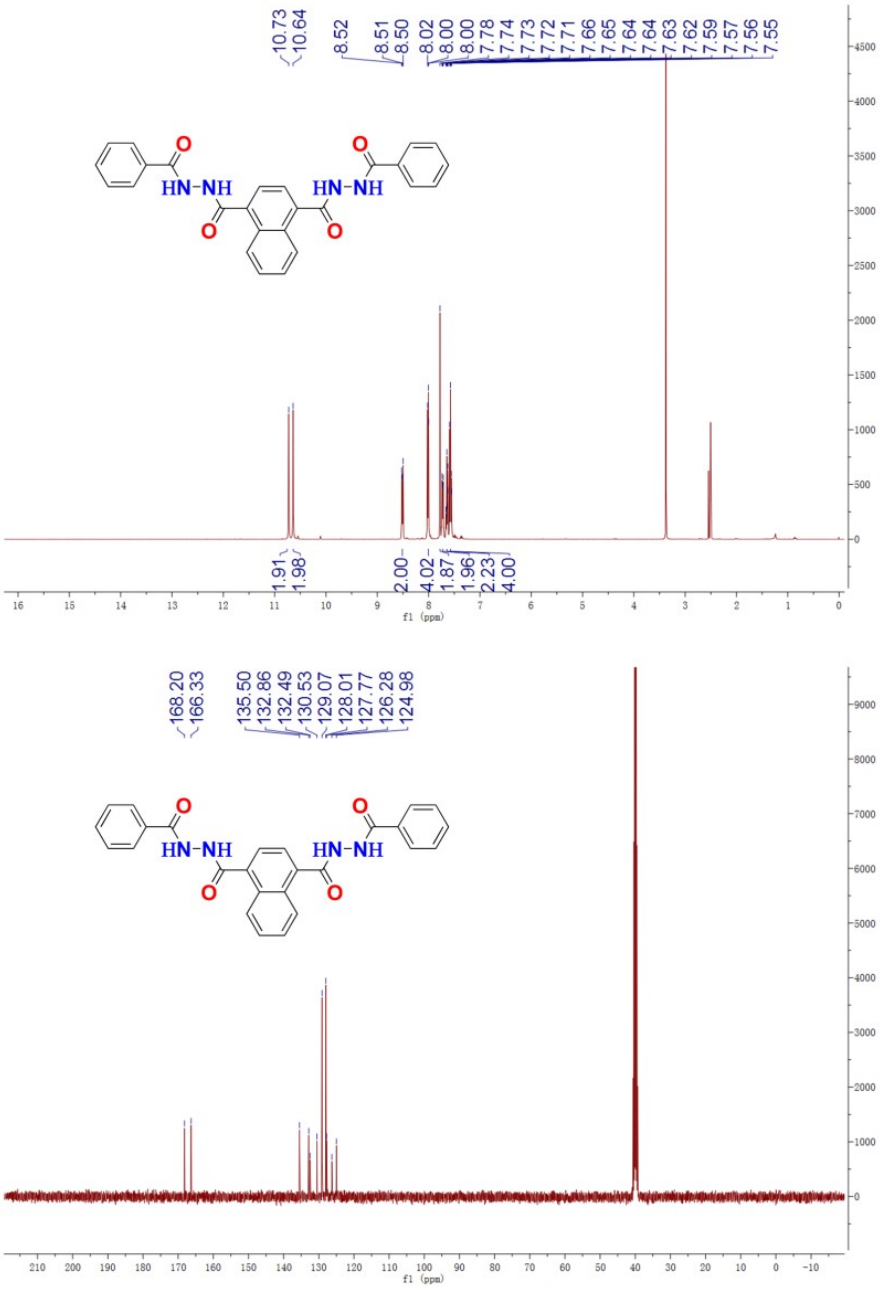


Fig. S2. ^{1}H NMR and ^{13}C NMR spectra of NDC-NNpH in DMSO-*d*6

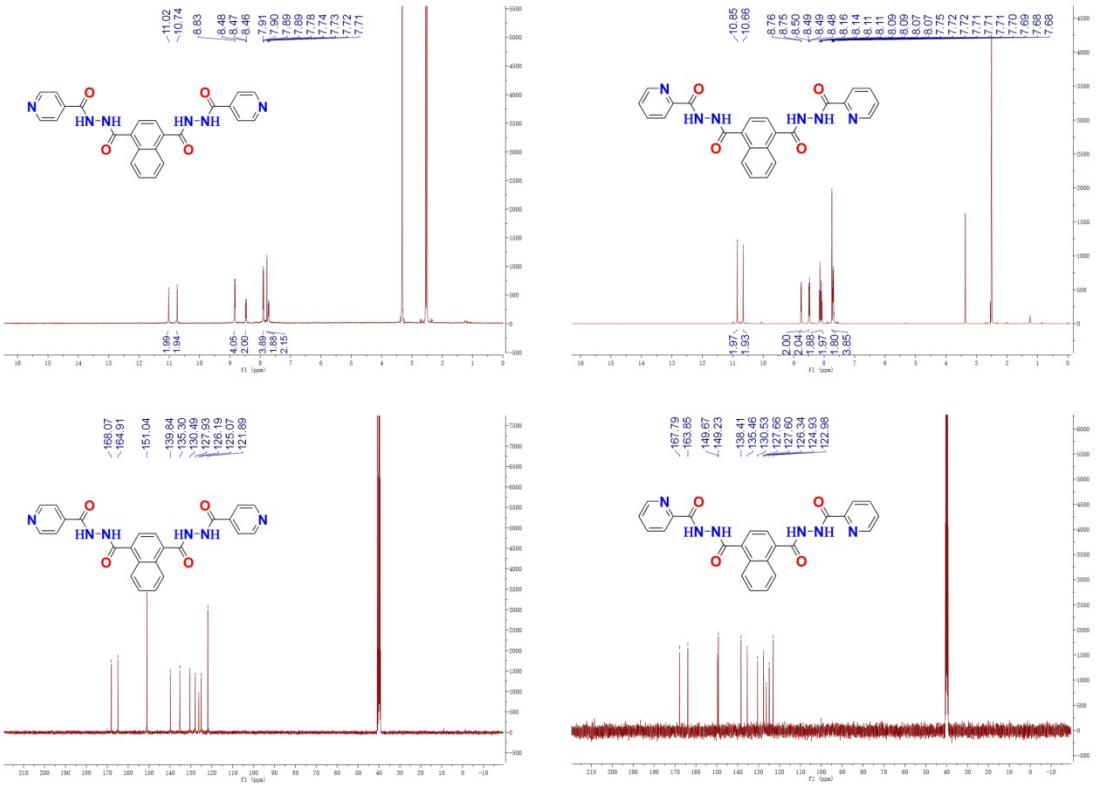


Fig. S3. ^{1}H NMR and ^{13}C NMR spectra of NDC-NN4/NDC-NN2 in DMSO-*d*6

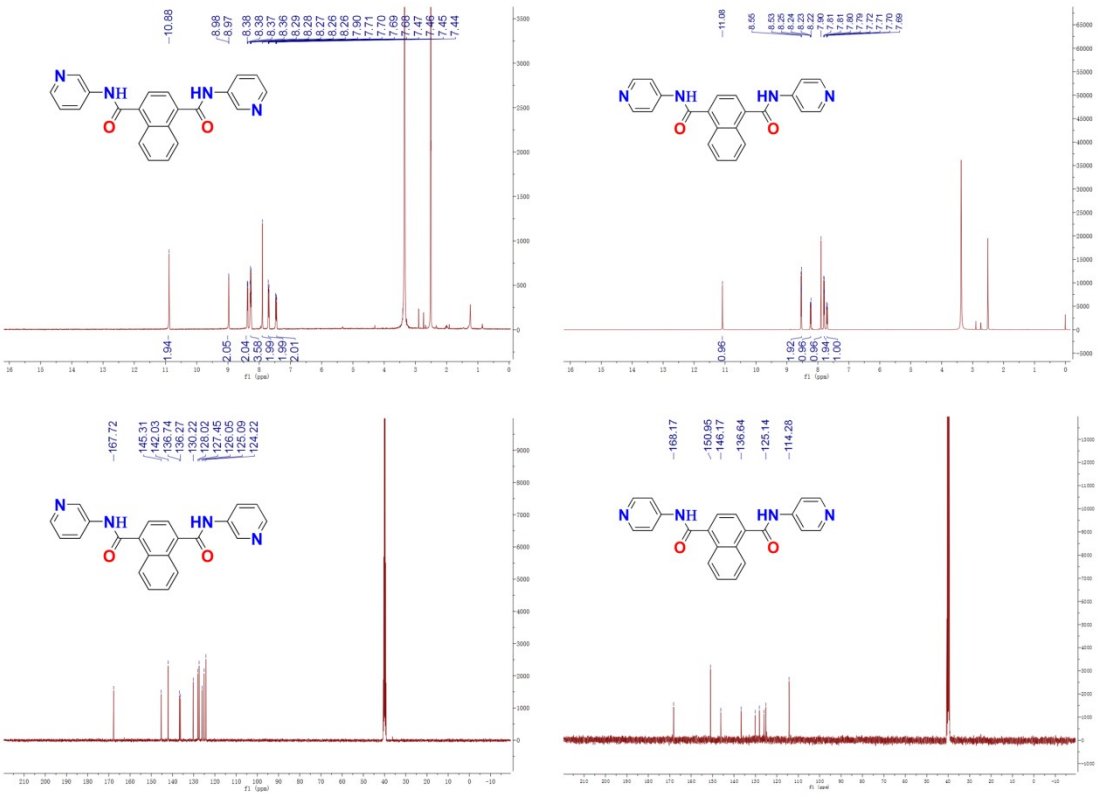


Fig. S4 ^{1}H NMR and ^{13}C NMR spectra of NDC-N3/NDC-N4 in DMSO-*d*6

Table S1. Gelation behavior of NDC-NN3, NDC-NN4 and NDC-NN2 under sonication in various solvents

	Solvents	Compounds		
		NDC-NN3	**NDC-NN4**	**NDC-NN2**
1	Methanol	S (S)	S (S)	P (P)
2	Ethanol	G (S)	S (S)	P (P)
3	*n*-Propanol	G (S)	S (S)	I (I)
4	*n*-Butanol	G (S)	S (S)	I (I)
5	*n*-Pentanol	G (S)	S (S)	P (P)
6	THF	G (S)	S (S)	S (S)
7	1,4-Dioxane	G (S)	S (S)	P (P)
8	Ethylene glycol	S (S)	S (S)	P (S)
9	H_2O	I (I)	I (I)	I (I)
10	DMSO	S (S)	S (S)	S (S)
11	DMF	S (S)	S (S)	S (S)
12	Aminobenzene	S (S)	S (S)	S (S)
13	Acetonitrile	I (I)	S (S)	I (I)
14	Pyridine	S (S)	S (S)	S (S)
15	Acetone	I (I)	S (S)	P (P)
16	$CHCl_3$	I (I)	I (I)	P (P)
17	Ethyl acetate	I (I)	I (I)	I (I)
18	Tetralin	P (P)	I (I)	I (I)
19	*n*-Butyl acetate	I (I)	I (I)	I (I)
20	CH_2Cl_2	P (P)	I (I)	I (I)
21	Benzene	P (P)	I (I)	I (I)
22	Ethyl ether	I (I)	I (I)	I (I)
23	*o*-Dichlorobenzene	I (I)	I (I)	I (I)
24	Chlorobenzene	I (I)	I (I)	I (I)
25	Mesitylene	I (I)	I (I)	I (I)
26	*p*-Xylene	I (I)	I (I)	I (I)
27	Toluene	I (I)	I (I)	I (I)
28	Isopropyl ether	I (I)	I (I)	I (I)
29	CCl_4	I (I)	I (I)	I (I)
30	Cyclohexane	I (I)	I (I)	I (I)
31	Hexane	I (I)	I (I)	I (I)
32	Petroleum ether	I (I)	I (I)	I (I)

* G = gel; S = solution; P = precipitate; I = insoluble. Glass vials containing compounds and solvents were heated and then cooled to room temperature. Subsequently, the mixed solutions were sonicated (40 KHz, 0.40 $W \cdot cm^{-2}$) in deionized water for a certain time. After standing at room temperature for 24 h without sonication, the values in parentheses were recorded.

Table S2. Gelation behavior of NDC-NNpH, NDC-N3 and NDC-N4 under sonication in various solvents

	Solvents	Compounds		
		NDC-NNpH	**NDC-N3**	**NDC-N4**
1	Methanol	P (S)	P (P)	P (S)
2	Ethanol	I (I)	P (P)	P (S)
3	*n*-Propanol	I (I)	I (I)	P (S)
4	*n*-Butanol	I (I)	I (I)	I (I)
5	*n*-Pentanol	I (I)	I (I)	I (I)
6	THF	I (I)	I (I)	I (I)
7	1,4-Dioxane	I (I)	I (I)	I (I)
8	Ethylene glycol	P (S)	P (P)	I (S)
9	H_2O	I (I)	I (I)	I (I)
10	DMSO	S (S)	S (S)	P (S)
11	DMF	S (S)	S (S)	P (S)
12	Aminobenzene	S (S)	S (S)	P (S)
13	Acetonitrile	I (I)	I (I)	I (I)
14	Pyridine	I (I)	I (I)	I (I)
15	Acetone	I (I)	I (I)	I (I)
16	$CHCl_3$	I (I)	I (I)	I (I)
17	Ethyl acetate	I (I)	I (I)	I (I)
18	Tetralin	I (I)	I (I)	I (I)
19	*n*-Butyl acetate	I (I)	I (I)	I (I)
20	CH_2Cl_2	I (I)	I (I)	I (I)
21	Benzene	I (I)	I (I)	I (I)
22	Ethyl ether	I (I)	I (I)	I (I)
23	*o*-Dichlorobenzene	I (I)	I (I)	I (I)
24	Chlorobenzene	I (I)	I (I)	I (I)
25	Mesitylene	I (I)	I (I)	I (I)
26	*p*-Xylene	I (I)	I (I)	I (I)
27	Toluene	I (I)	I (I)	I (I)
28	Isopropyl ether	I (I)	I (I)	I (I)
29	CCl_4	I (I)	I (I)	I (I)
30	Cyclohexane	I (I)	I (I)	I (I)
31	Hexane	I (I)	I (I)	I (I)
32	Petroleum ether	I (I)	I (I)	I (I)

* G = gel; S = solution; P = precipitate; I = insoluble. Glass vials containing compounds and solvents were heated and then cooled to room temperature. Subsequently, the mixed solutions were sonicated (40 KHz, 0.40 $W \cdot cm^{-2}$) in deionized water for a certain time. After standing at room temperature for 24 h without sonication, the values in parentheses were recorded.

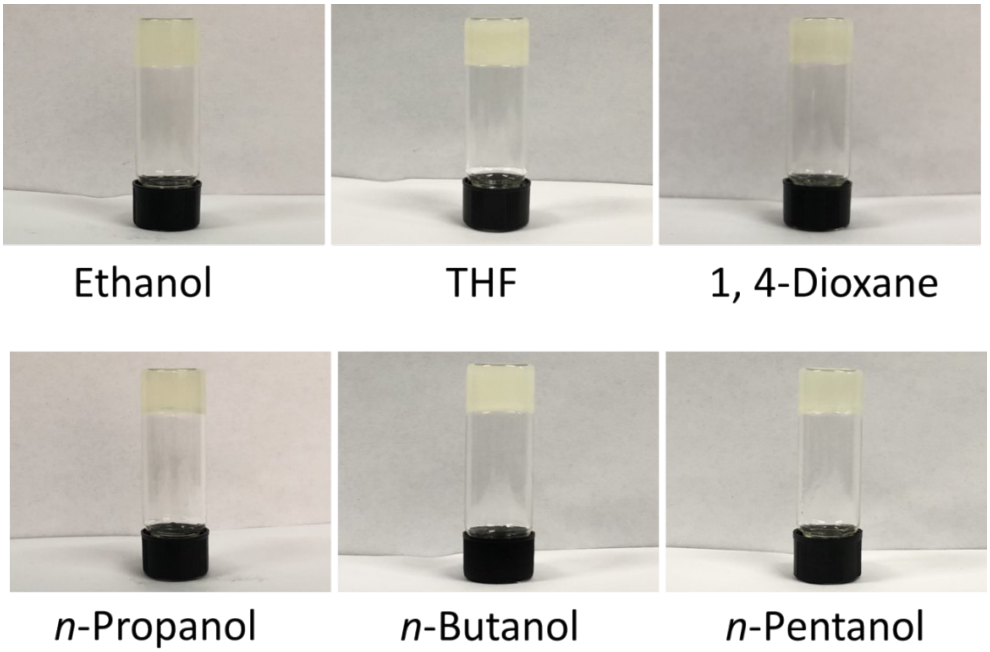


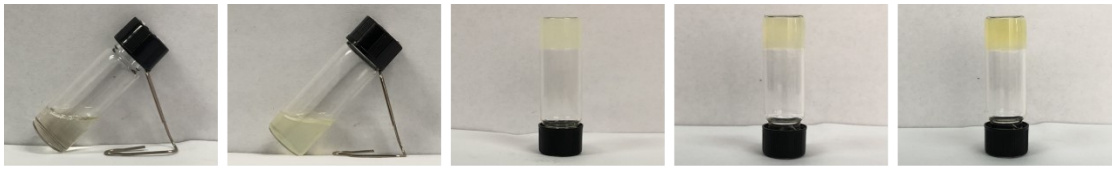
Fig. S5. Photographs of NDC-NN3 gels formed under ultrasonic irradiation in various solvents at 10 mg·mL^{-1}

Table S3. Gelation behavior of NDC-NN3 upon different ultrasonic time in ethanol, THF and 1, 4-dioxane with various concentrations

Solvents	$C_{NDC-NN3}$	Ultrasonic times / min										
	mg·mL^{-1}	1	2	5	10	15	20	25	30	40	60	120
Ethanol	6	S	S	S	S	S	S	S	S	S	S	S
	8	S	S	S	S	S	S	S	S	S	S	TS
	10	S	S	S	S	S	S	S	S	TS	G	G
	20	S	S	S	TS	TS	G	G	G	G	G	G
	30	S	TS	TS	G	G	G	G	G	G	G	G
	40	TS	TS	G	G	G	G	G	G	G	G	G
	45	I	/	/	/	/	/	/	/	/	/	/
THF	2	S	S	S	S	S	S	S	S	S	S	S
	4	S	S	S	S	S	S	S	S	S	S	S
	6	S	S	S	S	S	S	S	S	S	S	TS
	8	S	S	S	S	S	S	S	TS	TS	G	G
	10	S	S	S	TS	TS	TS	TS	G	G	G	G
	15	I	/	/	/	/	/	/	/	/	/	/
1,4-Dioxane	2	S	S	S	S	S	S	S	S	S	S	S
	4	S	S	S	S	S	P	TS	TS	TS	TS	TS
	6	S	S	S	TS	TS	G	G	G	G	G	G
	8	S	S	TS	G	G	G	G	G	G	G	G
	10	S	TS	G	G	G	G	G	G	G	G	G
	15	I	/	/	/	/	/	/	/	/	/	/
***n*-Propanol**	4	S	S	S	S	S	S	S	S	S	S	S
	6	S	S	S	S	S	S	S	S	S	S	TS
	8	S	S	S	S	S	S	S	S	TS	TS	G
	10	S	S	S	S	S	S	S	S	TS	G	G
	15	S	S	S	TS	TS	TS	TS	G	G	G	G
	20	I	/	/	/	/	/	/	/	/	/	/

***n*-Butanol**	2	S	S	S	S	S	S	S	S	S	S	S
	4	S	S	S	S	S	S	S	S	S	TS	TS
	6	S	S	S	S	S	TS	TS	G	G	G	G
	8	S	S	S	S	TS	TS	G	G	G	G	G
	10	S	S	TS	TS	TS	G	G	G	G	G	G
	15	I	/	/	/	/	/	/	/	/	/	/
***n*-Pentanol**	2	S	S	S	S	S	S	S	S	S	S	S
	4	S	S	S	S	S	S	S	S	S	S	S
	6	S	S	S	S	S	S	S	S	S	S	TS
	8	S	S	S	S	S	S	S	S	TS	TS	G
	10	S	S	S	S	S	TS	TS	TS	TS	G	G
	15	I	/	/	/	/	/	/	/	/	/	/

* G = gel; I = insoluble partially; S = solution; TS = turbid solution.



6 mg·mL^{-1} (0.76 wt.%) | 8 mg·mL^{-1} (1.01 wt.%) | 10 mg·mL^{-1} (1.27 wt.%) | 20 mg·mL^{-1} (2.53 wt.%) | 40 mg·mL^{-1} (5.07 wt.%)

Fig. S6. Images of NDC-NN3 sonogels formed under ultrasonic irradiation in ethanol at different concentrations

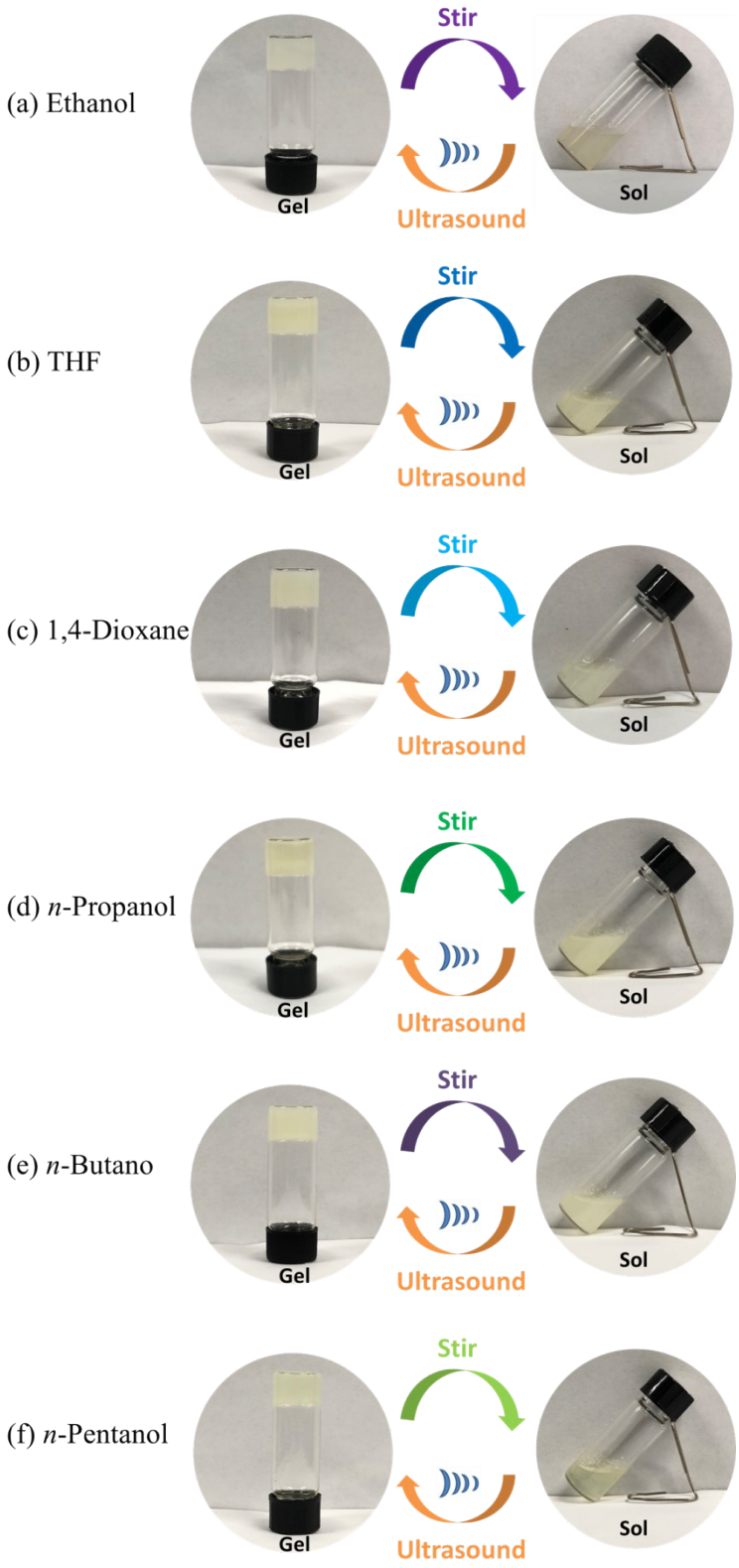


Fig. S7. Schematic diagrams of the reversible sol-gel transition processes for NDC-NN3 in different solvents (10 mg·mL^{-1}) under stirring and sonication: (a) ethanol; (b) THF; (c) 1,4-dioxane; (d) n-propanol; (e) n-butanol; (f) n-pentanol

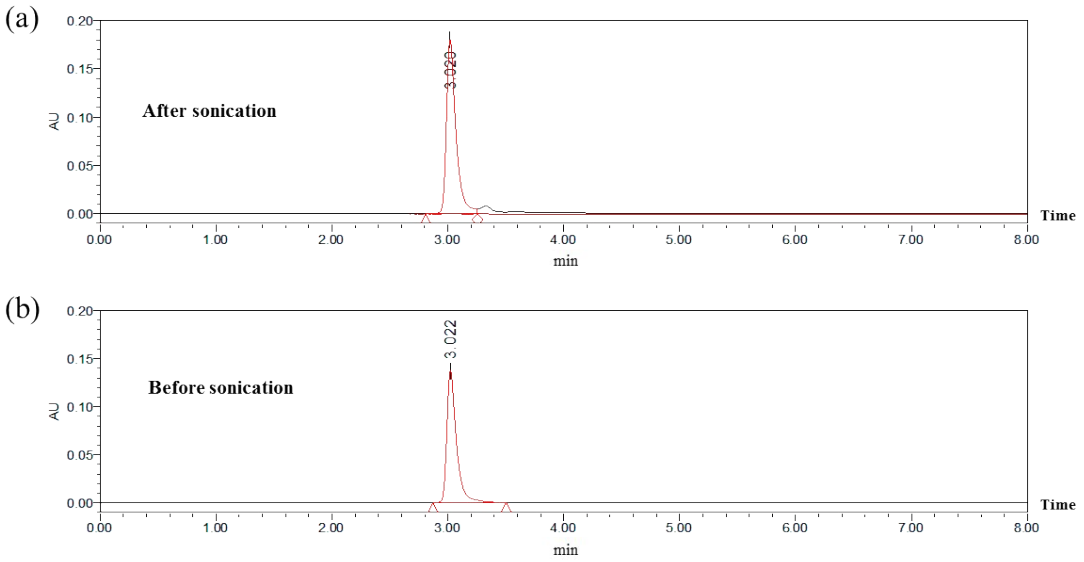


Fig. S8. HPLC chromatogram of gelator NDC-NN3: (a) after and (b) before formation of sonogel with CH_3OH as solution

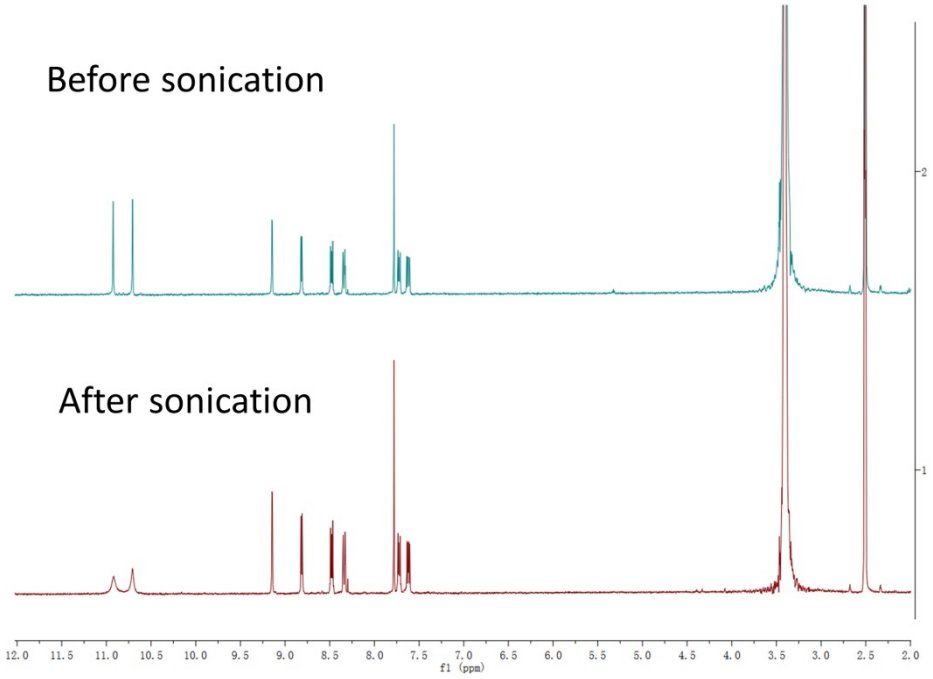


Fig. S9. ^{1}H NMR spectra of NDC-NN3 before and after sonication

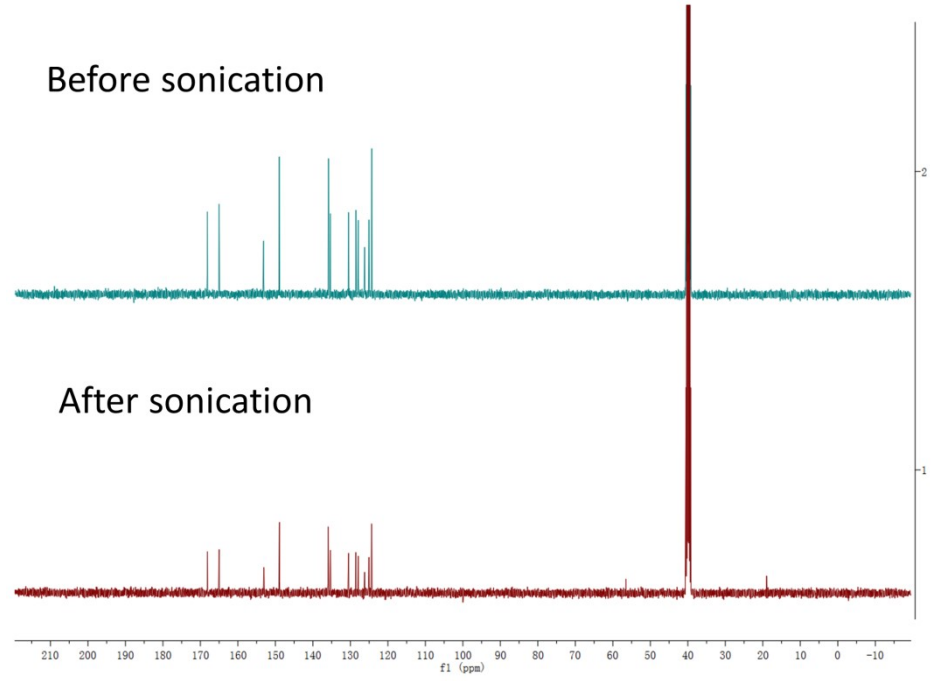


Fig. S10. ^{13}C NMR spectra of NDC-NN3 before and after sonication

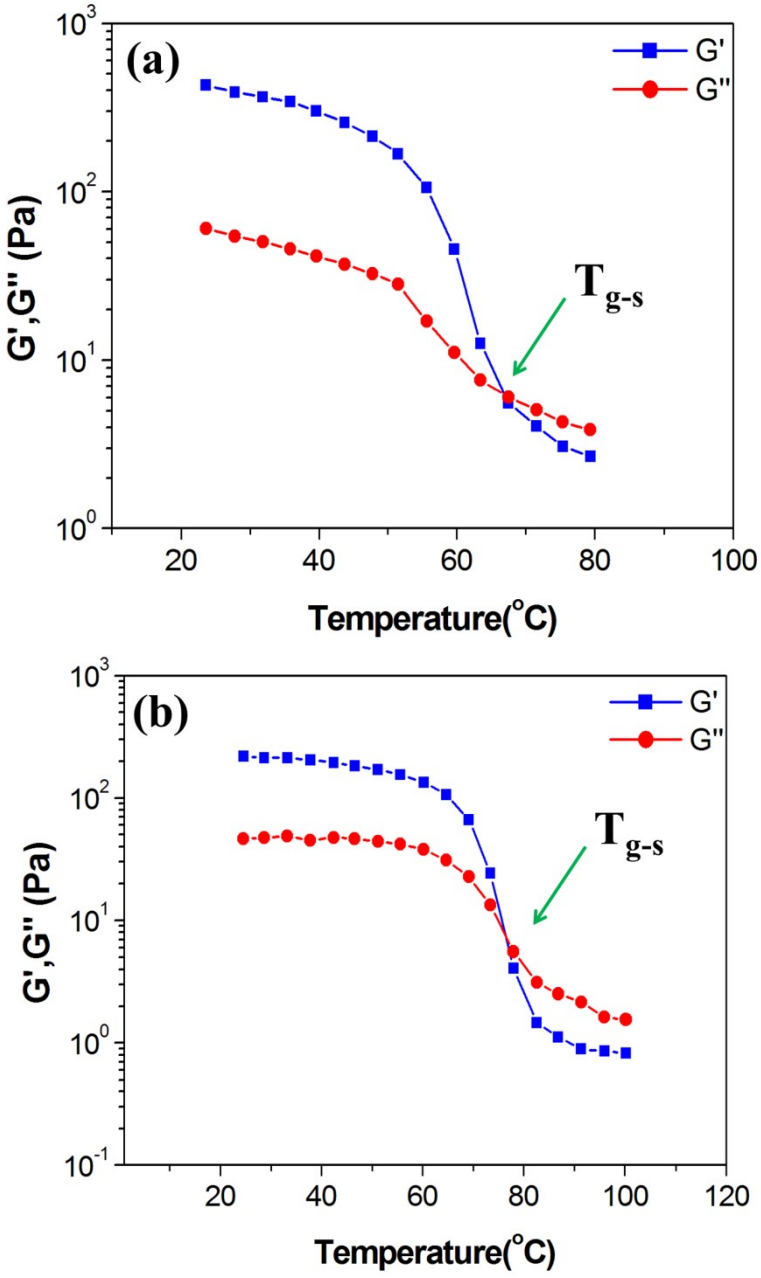


Fig. S11. Moduli variations of NDC-NN3 sonogels (10 mg·mL^{-1}) with increasing temperature: (a) ethanol and (b) 1,4-dioxane as solutions, respectively

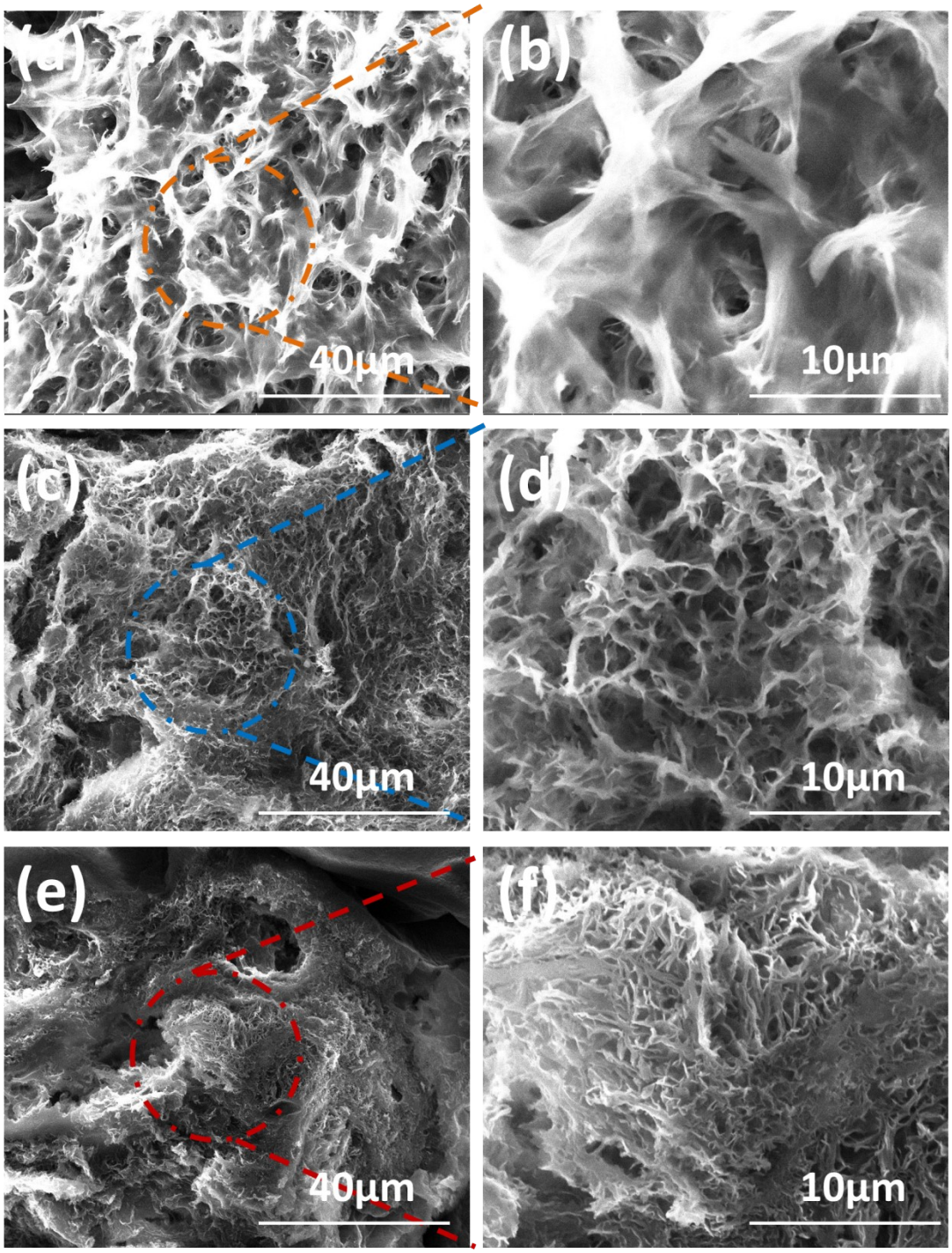


Fig. S12. SEM images of NDC-NN3 aggregates in dried sonogels formed in different solvents: (a,b) *n*-propanol; (c,d) *n*-butanol; (e,f) *n*-pentanol

Table S4. Transformation of gel-crystal for NDC-NN3 in various solvents

Solvents	C_{PDA-N4} mg·mL^{-1}	Time / Days							
		1D	2D	5D	10D	15D	30D	60D	90D
Ethanol	10	G	G	G	G	G+C	G+C	C	C
	20	G	G	G	G+C	G+C	C	C	C
	30	G	G	G+C	G+C	C	C	C	C
	40	G+C	G+C	G+C	G+C	C	C	C	C
THF	8	G	G	G	G	G	G	G	G
	10	G	G	G	G	G	G	G	G
1,4-Dioxane	6	G	G	G	G	G	G	G	G
	8	G	G	G	G	G	G	G	G
	10	G	G	G	G	G	G	G	G
n-Propanol	8	G	G	G	G	G	G	G	G
	10	G	G	G	G	G	G	G	G
	15	G	G	G	G	G	G	G	G
n-Butanol	6	G	G	G	G	G	G	G	G
	8	G	G	G	G	G	G	G	G
	10	G	G	G	G	G	G	G	G
n-Pentanol	8	G	G	G	G	G	G	G	G
	10	G	G	G	G	G	G	G	G

* G = gel; C = crystal.

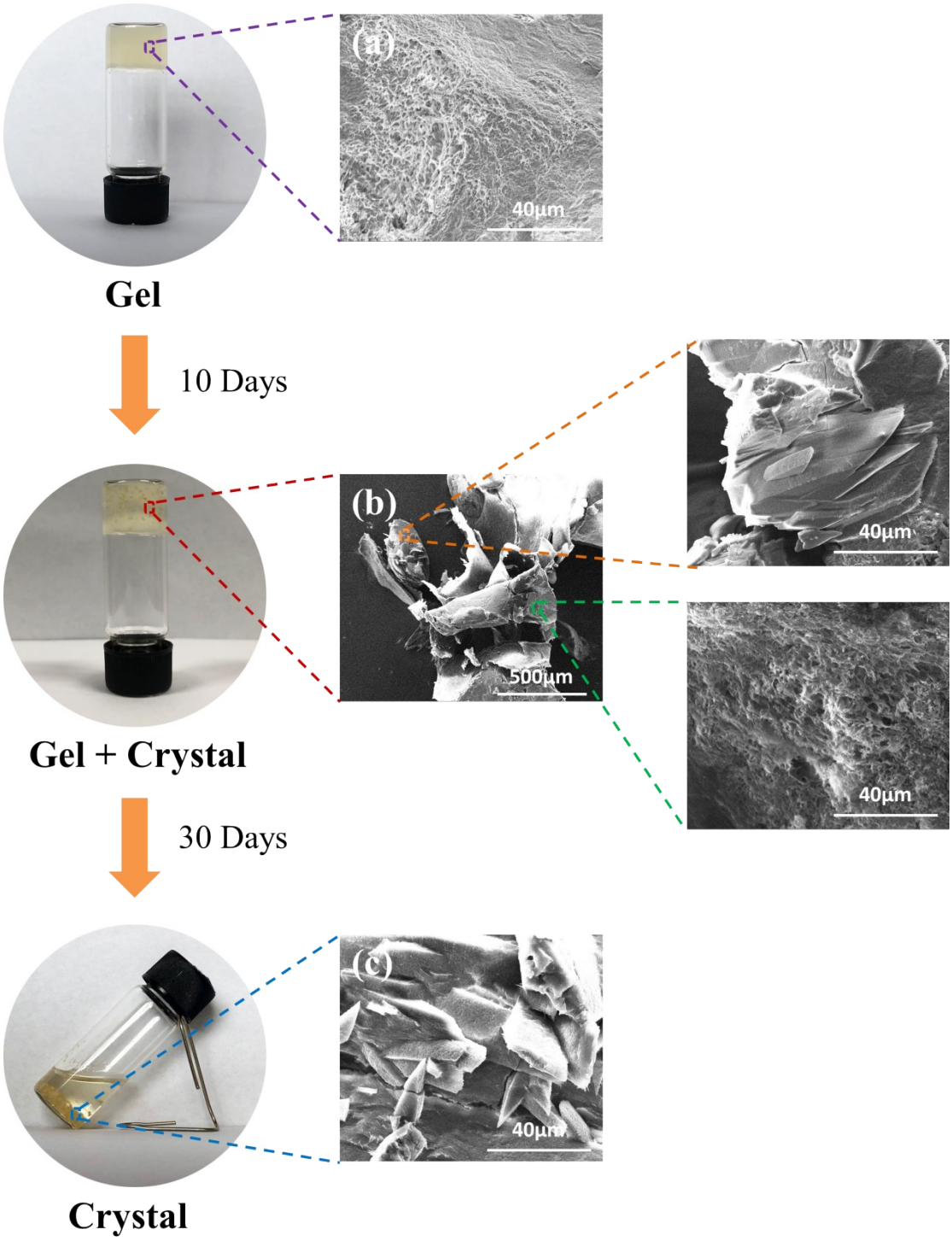


Fig. S13. Images of NDC-NN3 aggregates in sonogel (ethanol, 20 mg·mL^{-1}) aged at room temperature for (a) 0 day, (b) 10 days and (c) 30 days

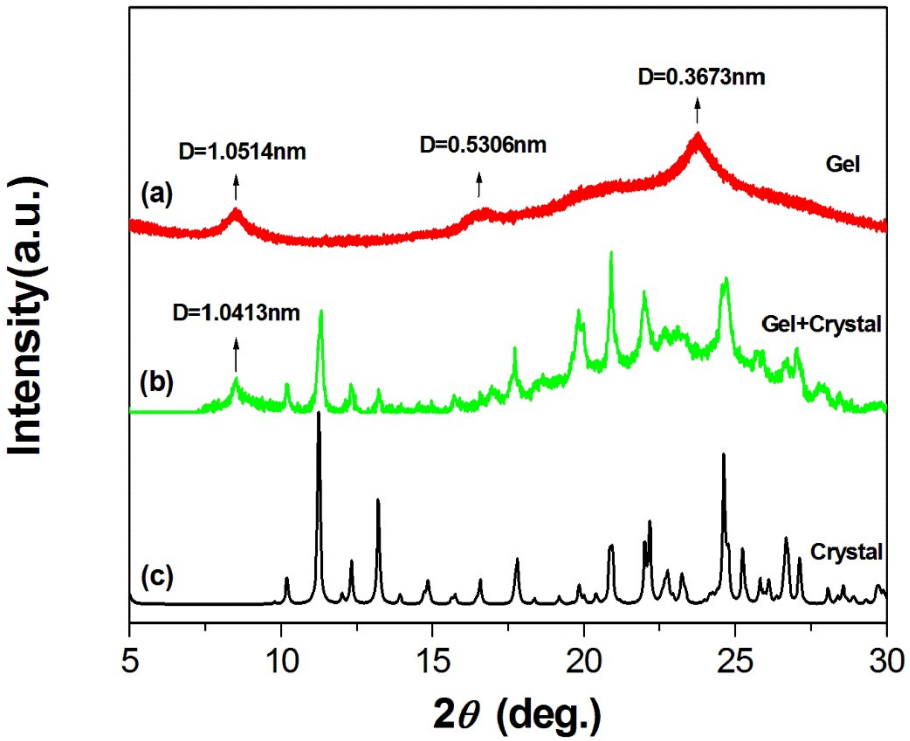


Fig. S14. SA-XRD patterns of NDC-NN3 sonogel (ethanol, 20 mg·mL^{-1}) aged at 25 °C for (a) 0 day, (b) 10 days and (c) 30 days (the simulated XRD pattern from the NDC-NN3 crystal data)

Table S5. Crystal data and structure refinement for NDC-NN3

Compound	**NDC-NN3**
Formula	$C_{102}H_{102}N_{24}O_{25}$
F.w.	2064.07
T **(K)**	100.01(10)
Crystal system	Monoclinic
Space group	$P21/n$
a (Å)	8.0569(2)
b (Å)	36.1507(7)
c (Å)	8.9337(2)
α (°)	90
β (°)	90.440(2)
γ (°)	90
V (Å^3)	2601.98(10)
Z	1
Dc (g/cm^3)	1.317
μ **(**mm^{-1}**)**	0.807
R_{int}	0.0782
GOF	1.050
$R1$, $wR2$ [$I > 2\sigma(I)$]	0.1023, 0.2832
$R1$, $wR2$ (all data)	0.1123, 0.2949

Table S6. Selected bond lengths (Å) and angles (º) for NDC-NN3 crystal

O4—C2	1.236(4)	N2—H2	0.8800
N6—C5	1.352(4)	N2—C20	1.376(7)
N6—C9	1.331(5)	N2—N1	1.377(5)
C2—N4	1.338(4)	C15—C18	1.373(6)
C2—C4	0.8800	C15—C20	1.507(5)
N3—N4	1.395(4)	C16—H16	0.9500
N3—C7	1.354(4)	C16—C17	1.411(6)
N4—H4	0.8800	C17—H17	0.9500
O3—C7	1.220(4)	C18—H18	0.9500
C3—H3A	0.9500	C19—N1	1.327(6)
C3—C4	1.386(5)	C19—C21	1.499(6)
C3—C6	1.388(5)	N1—H1	0.8800
C4—C5	1.384(5)	C21—C22	1.402(7)
C5—H5	0.9500	C21—C25	1.371(7)
C6—H6	0.9500	C22—H22	0.9500
C6—C9	1.378(5)	C22—N5	1.327(6)
C7—C11	1.509(4)	N5—C26	1.316(8)
C8—C10	1.435(5)	C24—H24	0.9500
C8—C11	1.428(5)	C24—C25	1.423(8)
C8—C12	1.406(6)	C24—C26	1.354(10)
C9—H9	0.9500	C25—H25	0.9500
C10—C14	1.438(6)	O5—H5A	0.8698
C10—C15	1.411(6)	O5—H5B	0.8707
C11—C13	1.353(6)	O8—H8	0.8400
O2—C20	1.221(6)	O8—C27	1.353(9)
O1—C19	1.312(5)	O6—H6A	0.8707
C12—H12	0.9500	O6—H6B	0.8698
C12—C16	1.373(6)	C27—H27A	0.9900

C13—H13	0.9500	C27—H27B	0.9900
C13—C18	1.422(5)	C27—C28	1.518(11)
C14—H14	0.9500	C28—H28A	0.9800
C14—C17	1.351(7)	C28—H28B	0.9800

C9—N6—C5	117.2(3)	N1—N2—H2	120.1
O4—C2—N4	123.7(3)	C10—C15—C20	119.2(4)
O4—C2—C4	121.8(3)	C18—C15—C10	121.9(3)
N4—C2—C4	114.5(3)	C18—C15—C20	118.7(4)
N4—N3—H3	121.2	C12—C16—H16	120.0
C7—N3—H3	121.2	C12—C16—C17	120.1(4)
C7—N3—N4	117.5(3)	C17—C16—H16	120.0
C2—N4—N3	120.6(3)	C14—C17—C16	120.1(4)
C9—N6—C5	117.2(3)	C14—C17—H17	120.0
C2—N4—H3	119.7	C16—C17—H17	120.0
N3—N4—H4	119.7	C13—C18—H18	120.3
C4—C3—H3A	120.8	C15—C18—C13	119.5(4)
C4—C3—C6	118.4(3)	C15—C18—H18	120.3
C6—C3—H3A	120.8	N1—C19—C21	116.3(4)
C3—C4—C2	123.9(3)	N2—C20—C15	112.5(4)
C5—C4—C2	117.3(3)	N2—N1—H1	119.4
C5—C4—C3	118.7(3)	C19—N1—N2	121.3(4)
N6—C5—C4	123.1(3)	C19—N1—H1	119.4
N6—C5—C4	118.5	C22—C21—C19	123.7(4)
C4—C5—H5	118.5	C25—C21—C19	119.5(5)
C3—C6—H6	120.5	C25—C21—C22	116.8(4)
C9—C6—C3	119.0(3)	C21—C22—H22	117.0
C9—C6—H6	120.5	N5—C22—C21	126.0(5)
N3—C7—C11	113.3(3)	N5—C22—H22	117.0

O3—C7—N3	123.7(3)	C26—N5—C22	115.4(5)
O3—C7—C11	122.9(3)	C25—C24—H24	120.6
C11—C8—C10	118.4(4)	C26—C24—H24	120.6
C12—C8—C10	119.3(3)	C26—C24—C25	118.8(5)
C12—C8—C11	122.3(3)	C21—C25—C24	117.8(6)
N6—C9—C6	123.5(3)	C21—C25—H25	121.1
N6—C9—H9	118.2	C24—C25—H25	121.1
C6—C9—H9	118.2	N5—C26—C24	125.2(5)
C8—C10—C14	117.0(4)	N5—C26—H26	117.4
C15—C10—C8	118.3(4)	C24—C26—H26	117.4
C15—C10—C14	124.7(4)	H5A—O5—H5B	104.4
C8—C11—C7	119.0(3)	C27—O8—H8	109.5
C13—C11—C7	119.5(3)	H6A—O6—H6B	104.5
C13—C11—C8	121.5(3)	O8—C27—H27A	107.4
C8—C12—H12	119.3	O8—C27—H27B	107.4
C16—C12—C8	121.4(4)	O8—C27—C28	119.8(4)
C16—C12—H12	119.3	H27A—C27—H27B	106.9
C11—C13—H13	119.8	C28—C27—H27A	107.4
C11—C13—C18	120.4(4)	C28—C27—H27B	107.4
C18—C13—H13	119.8	C27—C28—H28A	109.5
C10—C14—H14	118.9	C27—C28—H28B	109.5
C17—C14—C10	122.4(4)	C27—C28—H28C	109.5
C17—C14—H14	118.9	H28A—C28—H28B	109.5
C20—N2—H2	120.1	H28A—C28—H28C	109.5
C20—N2—N1	119.7(4)	H28B—C28—H28C	109.5

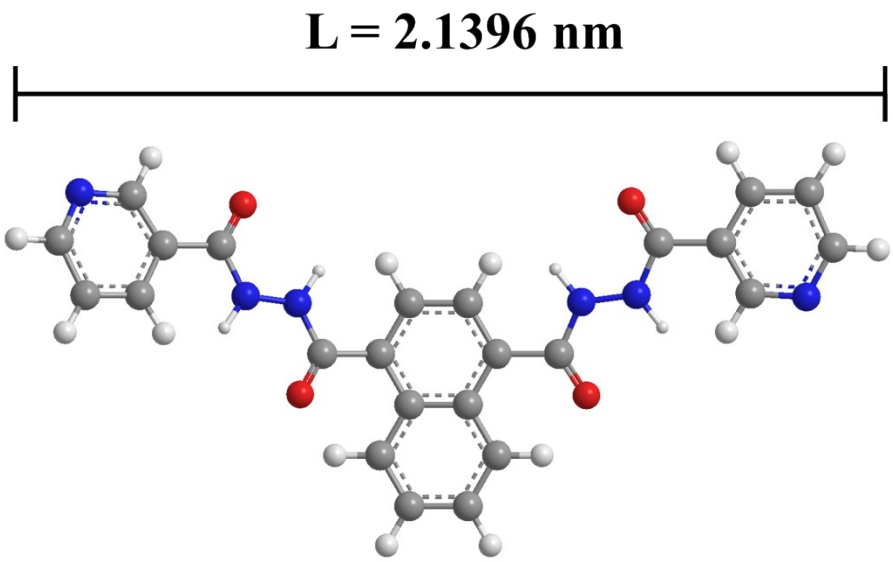


Fig. S15. CPK space-filling model of NDC-NN3

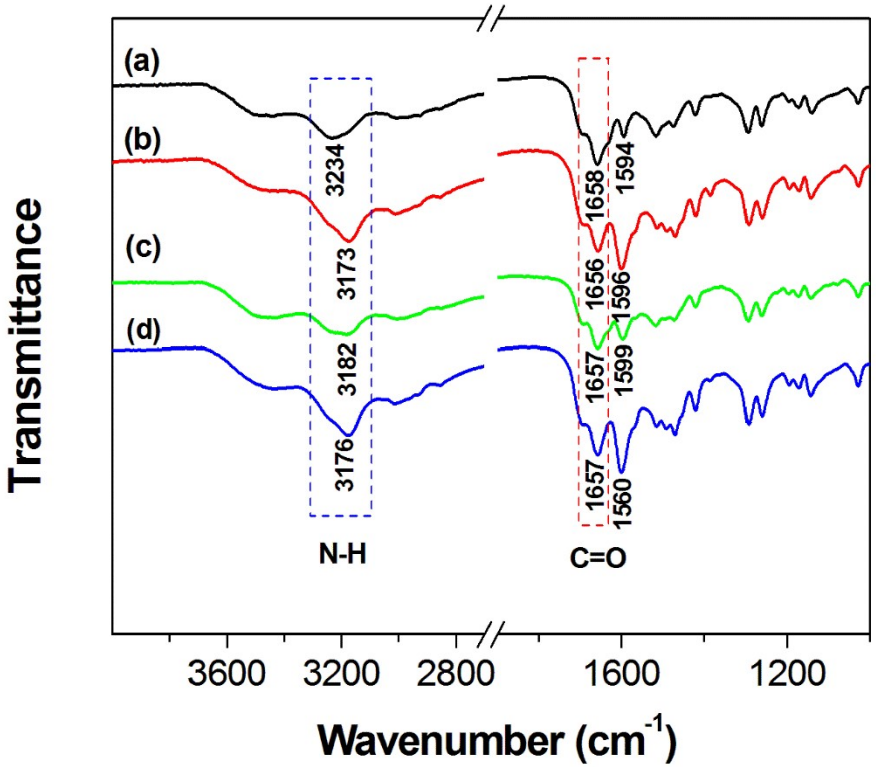


Fig. S16. FT-IR spectra of (a) NDC-NN3 crystal and xerogels formed from sonogels with (b) ethanol, (c) THF and (d) 1,4-dioxane as solvents

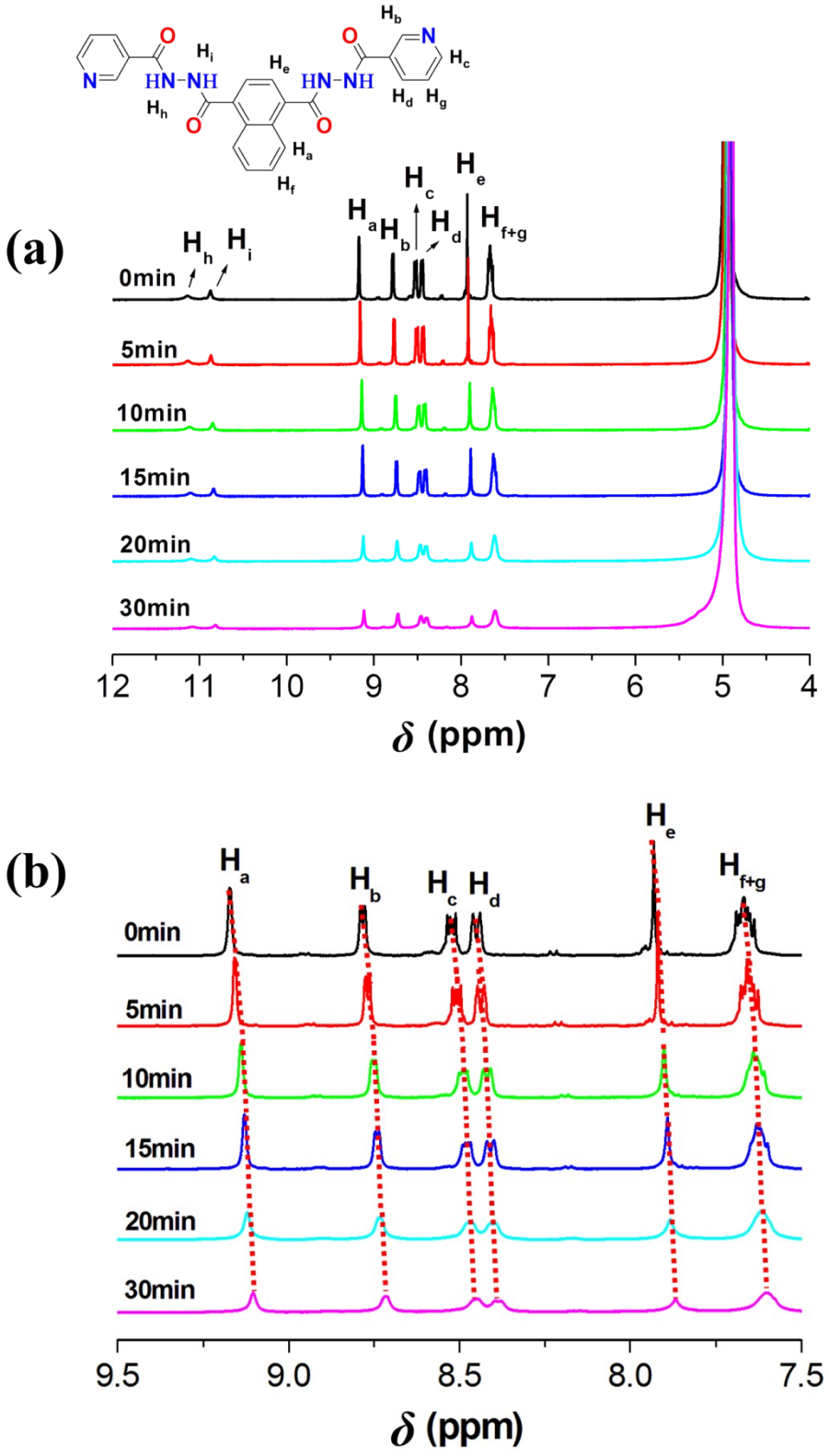


Fig. S17. (a) Variable-time ^{1}H NMR spectra of NDC-NN3 sonogel (ethanol-*d*6, 20 mg·mL^{-1}) and (b) the partial image enlargement

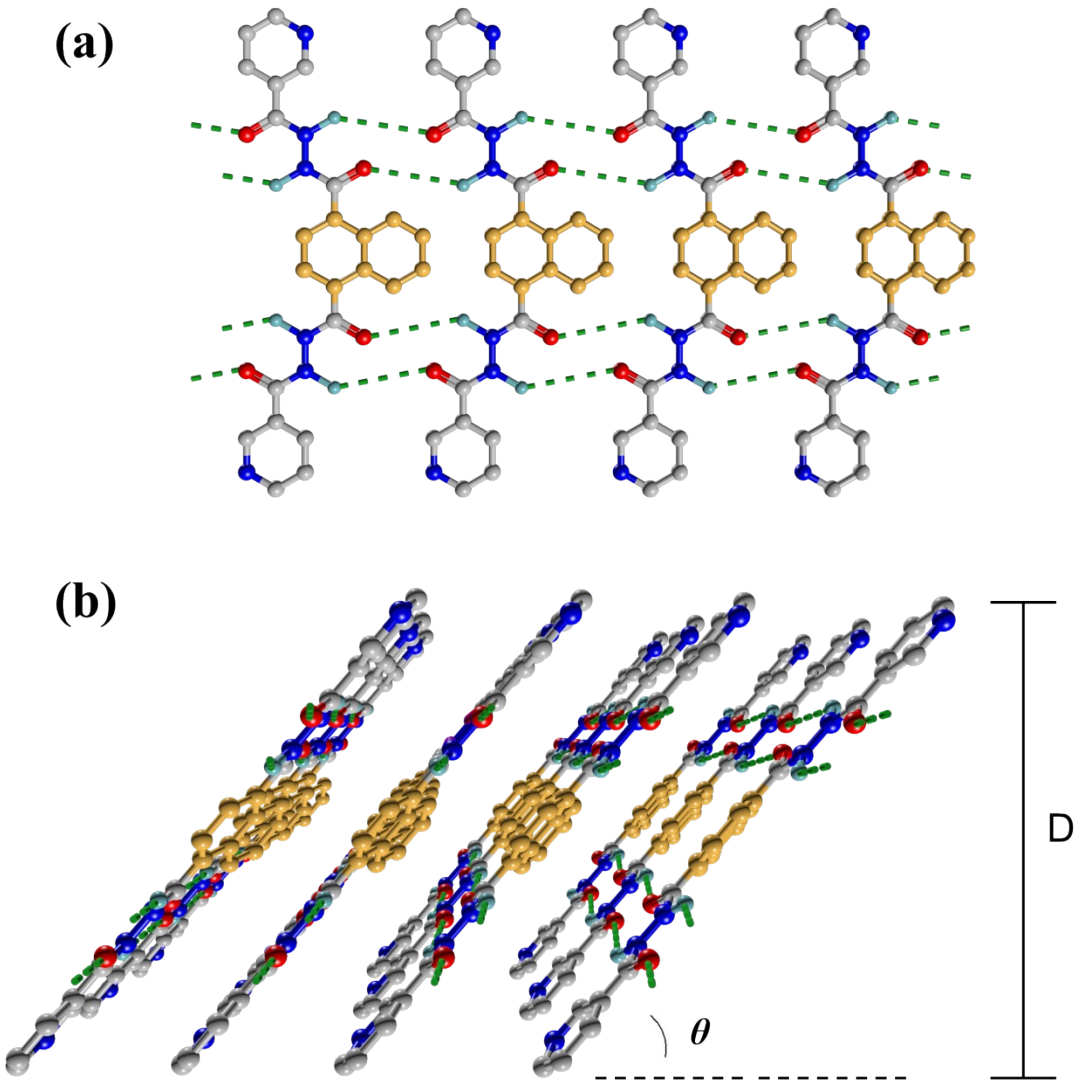


Fig. S18. Schematic representation of the possible local aggregation for NDC-NN3 in sonogels: (a) 2D and (b) 3D structure

2. References

1 X. Z. Luo, X. J. Jia, J. H. Deng, J. L. Zhong, H. J. Liu, K. J. Wang and D. C. Zhong, *J. Am. Chem. Soc.*, 2013, 135, 11684-11687.