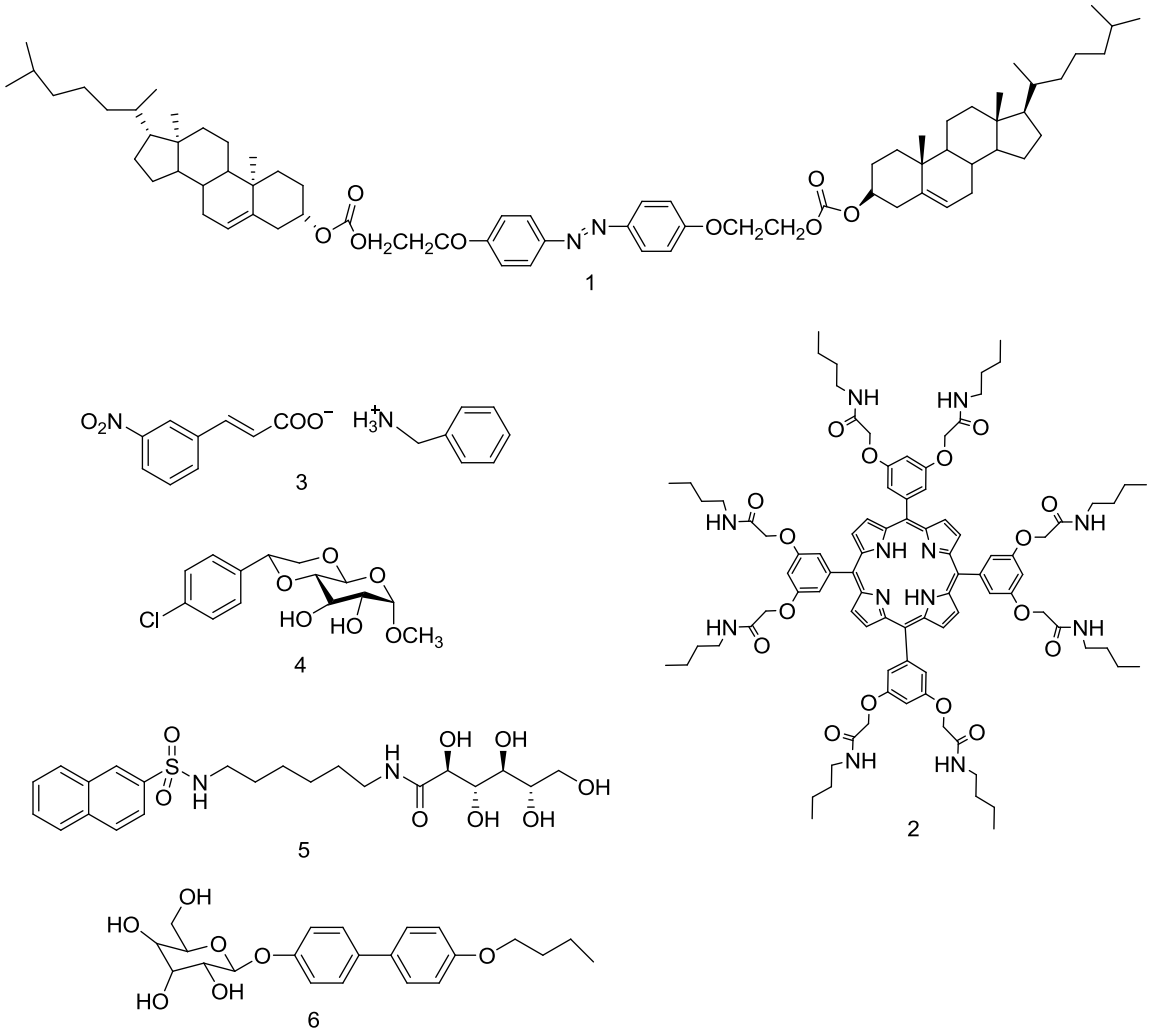
Supporting information

Application of the model to gelation behavior of the gelators reported in literatures[1-6]



Scheme S1 Structures of gelators **1, 2, 3, 4** ,**5** and **6** in literatures

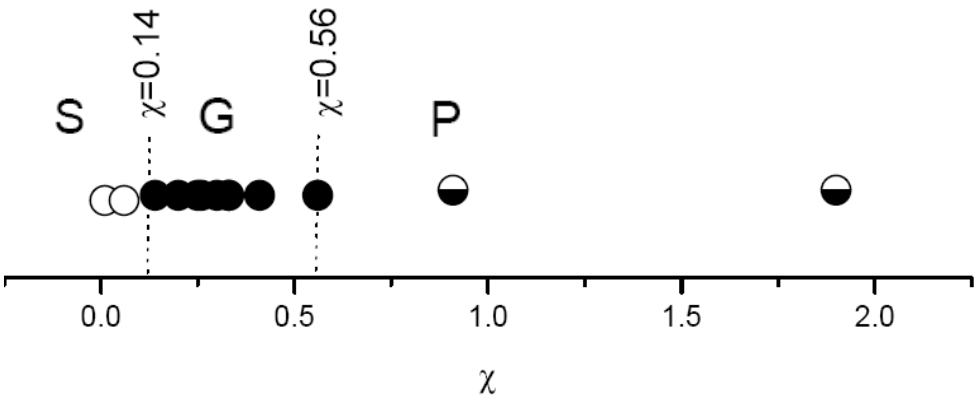


Figure S1. Calculated Flory-Huggins interaction parameter (χ) for gelator **1** (data taken from reference 1). S:solution, open circles; G: gelation, closed circles; P: precipitation, half closed circles.

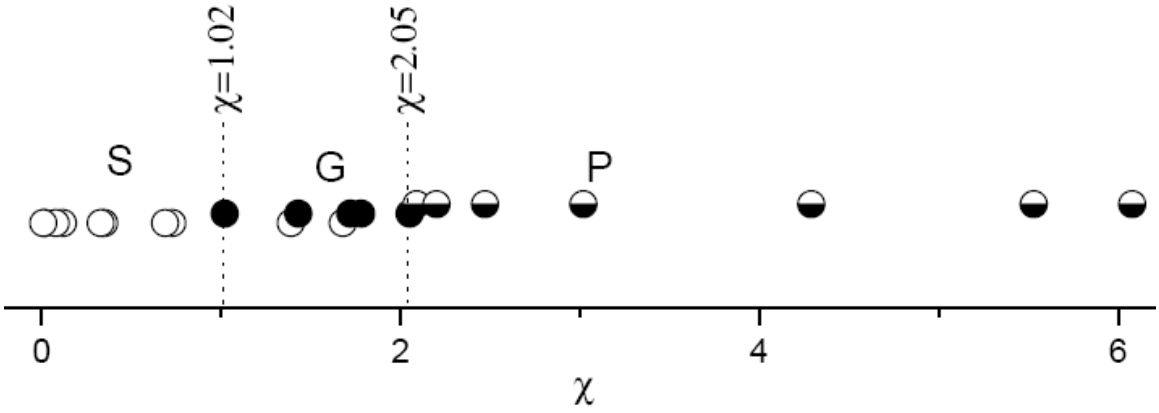


Figure S2. Calculated Flory-Huggins interaction parameter (χ) for gelator **2** (data taken from reference 2). S: solution, open circles; G: gelation, closed circles; P: precipitation, half closed circles.

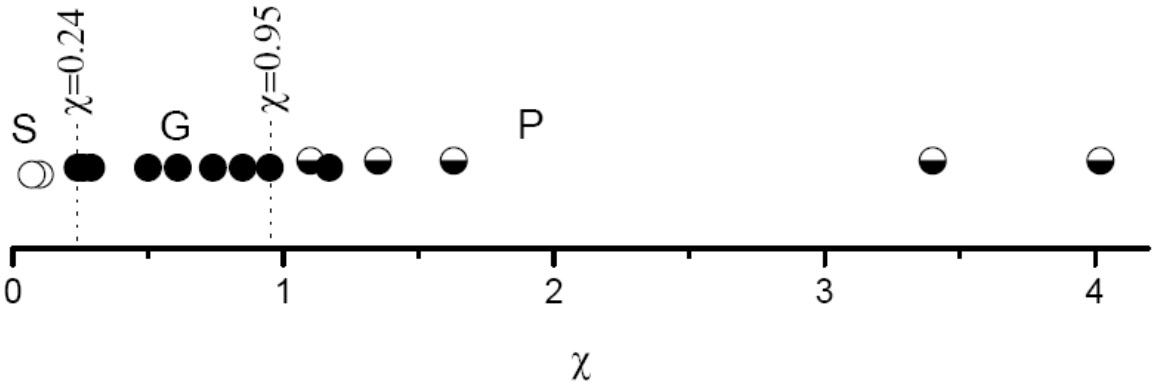


Figure S3. Calculated Flory-Huggins interaction parameter (χ) for gelator **3** (data taken from reference 3). S: solution, open circles; G: gelation, closed circles; P: precipitation, half closed circles.

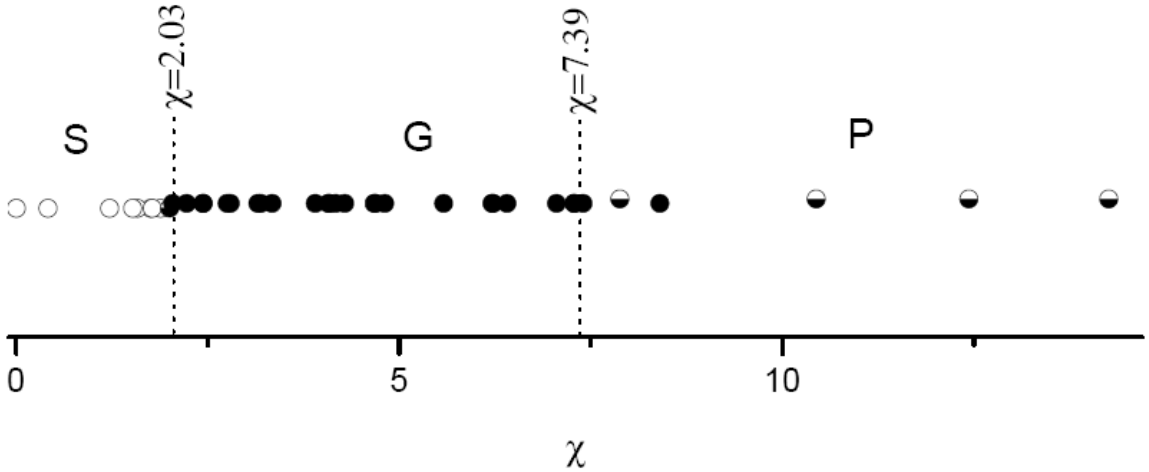


Figure S4. Calculated Flory-Huggins interaction parameter (χ) for gelator **4** (data taken from reference 4). S: solution, open circles; G: gelation, closed circles; P: precipitation, half closed circles.

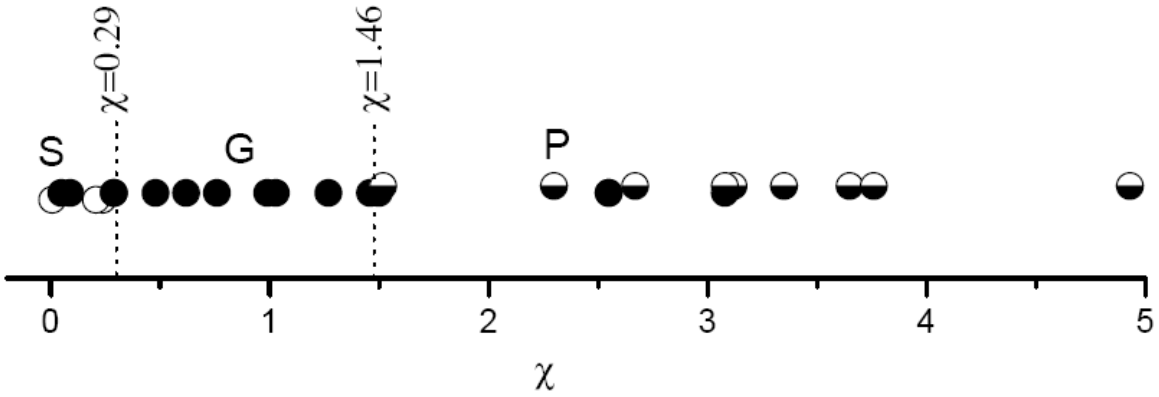


Figure S5. Calculated Flory-Huggins interaction parameter (χ) for gelator **5** (data taken from reference 5). S: solution, open circles; G: gelation, closed circles; P: precipitation, half closed circles.

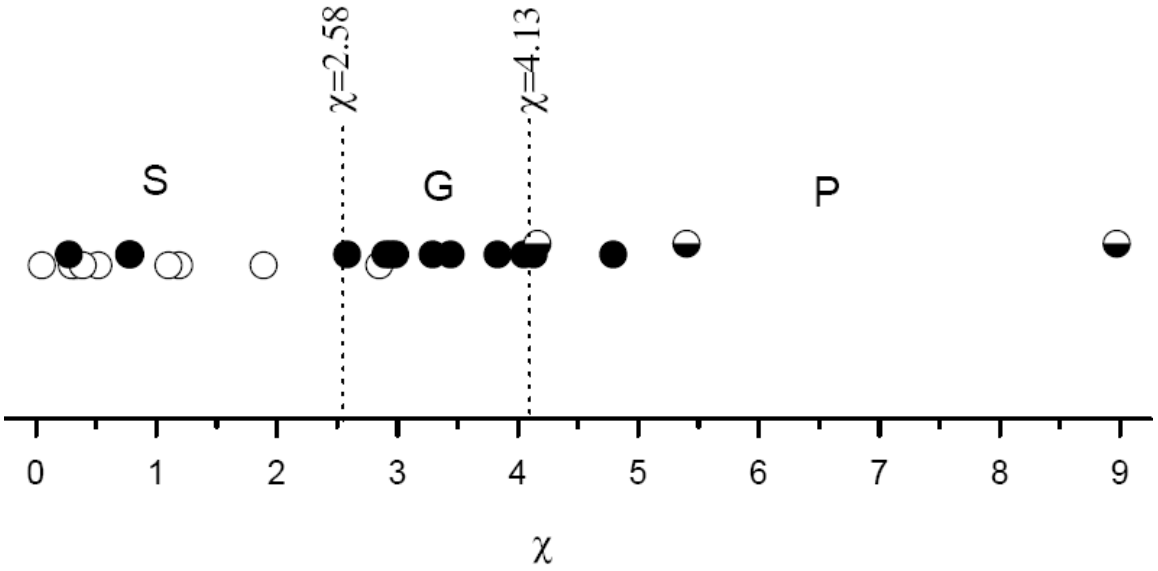


Figure S6. Calculated Flory-Huggins interaction parameter (χ) for gelator **6** (data taken from reference 6). S: solution, open circles; G: gelation, closed circles; P: precipitation, half closed circles.

Table S1. Duration of gel stability in different solvents.

[PM] (w/v)	duration of the gels				
	in H_2O	in MeCN-H_2O	in DMF-H_2O	in EtOH-H_2O	in EG-H_2O
1%	6 d	1 w	1 w	1 w	1 w
2%	2 d	1 w	1 w	1 w	1 w
3%	84 min	3 d	4 d	1 w	1 w
4%	28 min	24.2 h	23.3 h	5 d	25 h
5%	19 min	5.3 h	5.4 h	3 d	22.3 h
6%	12 min	1.2 h	4.3 h	18.0 h	3.7 h

MeCN-H_2O=acetonitrile-water, DMF-H_2O=dimethylformamide-water, EtOH-H_2O=ethanol-water, and EG-H_2O=ethylene glycol-water. The solvent mixtures contained 10 vol% organic solvent. The time started after the mixture immobility when the test tube was turned upside down. The detection period was one month.

Table S2 summarizes the solubility parameters and Flory-Huggins parameter (χ) to establish a model.

Table S2. Calculated solubility parameters (δ), molar volume (V) and Flory-Huggins parameter (χ) for PM and various solvents.

Solvents	$\delta(J^{1/2}/cm^{3/2})$	$V(cm^3/mol)$	χ	gelation behavior [a]
acetonitrile(MeCN)	24.3	52.6	0.16	S
ethanol(EtOH)	26.0	58.5	0.47	S
DMF	24.8	77.0	0.33	S
ethylene glycol(EG)	29.9	55.8	1.58	G
water	47.9	18.0	5.05	G-P
Dioxane (DO)	20.5	85.7	0.04	S
Methanol (MeOH)	29.7	40.7	1.10	S
DMSO	24.5	71.3	0.25	S
Glycerol (GI)	33.8	73.3	4.45	G
acetone	20.3	74	0.05	S
Isopropanol (IPA)	23.5	76.8	0.12	S
Ethoxyethanol (EE)	21.5	97.8	0.01	S
toluene	18.2	106.8	0.48	S
Cyclohexane (CyH)	16.8	108.7	0.98	S
MeCN:H_2O(v/v)				
90:10	26.8	44.1	0.47	S
80:20	29.0	38.0	0.86	S
70:30	31.5	33.4	1.32	G
60:40	33.8	29.7	1.80	G
50:50	36.2	26.8	2.31	G
40:60	38.5	24.4	2.84	G
30:70	40.9	22.4	3.39	G
20:80	43.3	20.7	3.94	G
10:90	45.6	19.3	4.52	G-P
EtOH:H_2O(v/v)				
90:10	28.4	47.8	0.86	S
80:20	30.4	40.3	1.28	S
70:30	32.7	34.9	1.73	G
60:40	34.8	30.8	2.19	G
50:50	37.0	27.5	2.65	G
40:60	39.2	24.9	3.14	G
30:70	41.4	22.7	3.62	G
20:80	43.6	20.9	4.11	G
10:90	45.8	19.3	4.59	G-P
DMF:H_2O(v/v)				
90:10	27.0	58.0	0.73	S
80:20	29.4	46.5	1.17	S
70:30	31.7	38.8	1.64	G
60:40	34.1	33.3	2.12	G
50:50	36.4	29.2	2.60	G
40:60	38.7	26.0	3.10	G
30:70	41.0	23.4	3.59	G
20:80	43.4	21.3	4.09	G
10:90	45.7	19.5	4.59	G-P
EG:H2O(v/v)				
90:10	31.7	46.1	1.93	G

80:20	33.5	39.3	2.28	G
70:30	35.3	34.2	2.63	G
60:40	37.1	30.3	2.98	G
50:50	39.0	27.2	3.33	G
40:60	40.8	24.7	3.68	G
30:70	42.6	22.6	4.04	G
20:80	44.4	20.8	4.38	G
10:90	46.2	19.3	4.73	G-P

One-component solubility parameter and molar volume are taken from reference 7. The solubility parameter values of the mixed solvents calculated according to $\delta=\Sigma\phi_i\delta_i$ (Equ. 68 of reference 7)

Table S3 summarizes Flory-Huggins parameter (χ) for various solvents to verify the model.

Table S3. Calculated solubility parameters and Flory-Huggins parameter (χ) for PM and various mixed solvents.

Solvents	δ ($J^{1/2}/cm^{3/2}$)	V(cm^3/mol)	χ	gelation behavior [a]
DO/H_2O(v/v=1/1)	34.2	23.8	1.54	G
DO/H_2O(v/v=1/9)	45.5	19.5	4.52	G-P
MeOH/H_2O(v/v=1/1)	37.1	22.7	2.22	G
MeOH/H_2O(v/v=1/9)	46.2	19.1	4.68	G-P
DMSO/H_2O(v/v=1/1)	37.2	23.3	2.31	G
DMSO/H_2O(v/v=1/9)	45.7	19.5	4.58	G-P
GI/H_2O(v/v=1/1)	40.9	28.9	4.37	G
GI/H_2O(v/v=1/9)	46.6	19.5	4.93	G-P
acetone/H_2O(v/v=1/1)	33.8	23.4	1.42	G
acetone/H_2O(v/v=1/9)	45.4	19.5	4.48	G-P
IPA/H_2O(v/v=1/1)	35.8	23.5	1.93	G
EE/H_2O(v/v=1/1)	35.7	24.2	1.96	G
MeOH/toluene(1/1)	22.2	46.7	0.01	S
DO/toluene(v/v=1/1)	22.2	56.5	0.01	S
MeOH/CyH(v/v=1/1)	21.5	47.0	0.01	S
DO/CyH(v/v=1/1)	21.5	56.9	0.01	S

[a] Concentration: 4% (w/v). S=solution; G=gel; G-P=gel-after-precipitation. The detection period was one month.

The solubility parameter values of the mixed solvents calculated according to $\delta=\Sigma\phi_i\delta_i$ (Equ. 68 of reference 7.)

Table S4 shows the group additivity values used in Fedors polymer solubility theories.[8] The value of $-PO_4$ for the anion $-PO_4^-$ and the value of N (tertiary amine) for the ammonium cation NH^+ were used. These are at best strong approximations and have been used in some references.[4, 9]

Table S4. Summary of Group Additivity Values for PM

Fedors Method								
Groups	$-CH_3$	$-CH_2$	$-CH<$	$-NH_2$	$-PO_3(O^-)$ [a]	$-C=N-$	$=N^+H-$ [b]	conjugated double bond
Occurrences, N_i	4	10	2	3	1	3	1	3
E_i(J/mol)	4710	4940	3430	12560	20930	18840	11720	1670
V_i(cm^3/mol)	33.5	16.1	-1	19.2	28	23.1	5	-2.2
Sum	$E_{coh}=\sum E_i N_i$ =206960 $V=\sum V_i N_i$ =446.3							
$\delta(J^{1/2}/cm^{3/2})$	$\delta=(E_{coh}/V)^{1/2}$ =21.53							

[a] the value of $-PO_4$ for $-PO_4^-$; [b] the value of $=N-$ for $=N^+H-$.

Table S5 shows Flory-Huggins parameter (χ) that will be illustrated by example.[7-8]

Table S5. Calculated Flory-Huggins parameter for PM and acetonitrile-water mixture.

Example: Estimate the Flory-Huggins parameter (χ) of gelator PM in 50 % of vol fraction in acetonitrile-water mixture.
Solution: the Hildebrand solubility parameter $\delta=\delta_{AcN}$* vol fraction $+\delta_{H2O}$*(1- vol fraction)=36.2 $J^{1/2}/cm^{3/2}$. The molar volume $V= V_{AcN}$*mol fraction $+V_{H2O}$*(1-mol fraction)=26.8 cm^3/mol. According to eq. (1) $\chi=V_1 (\delta_2 - \delta_1)^2/RT = 26.8*(21.53 – 36.2)^2/8.314/298.15=2.31$

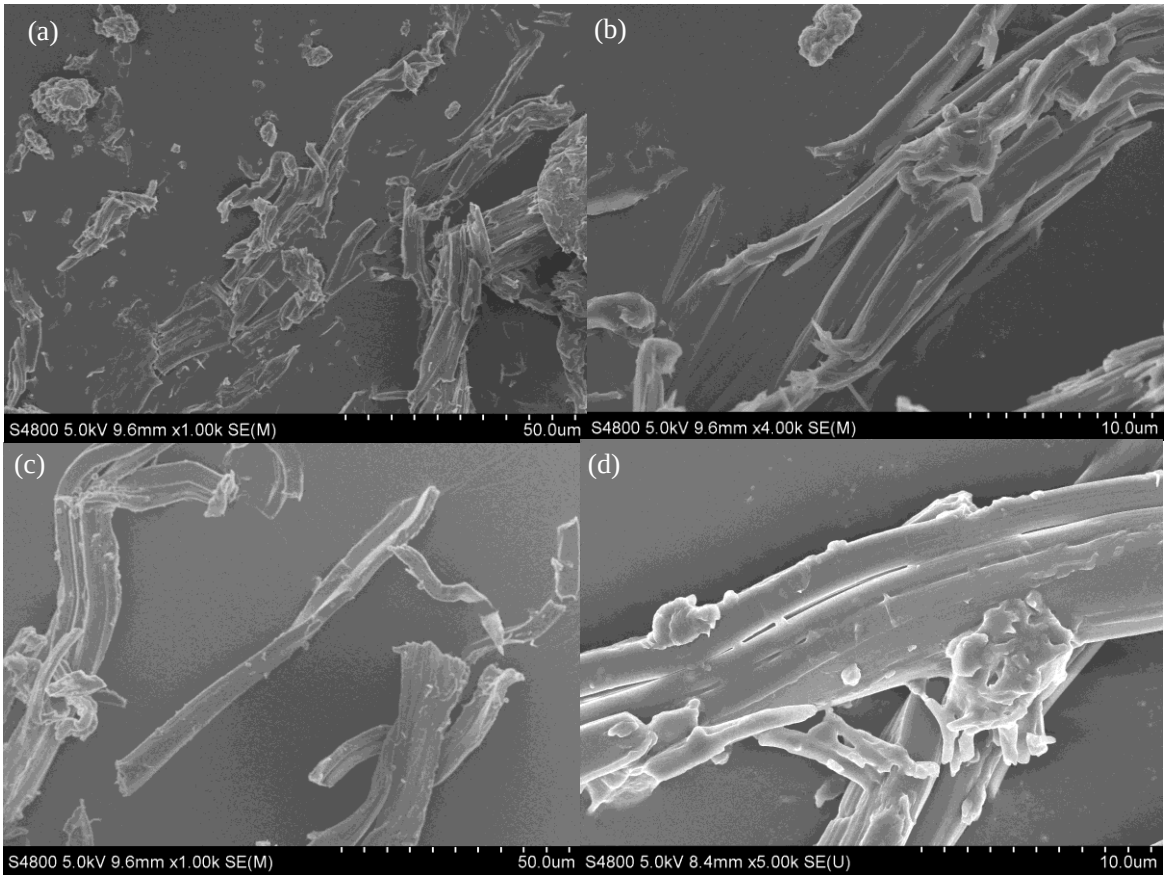


Figure S7. SEM micrographs of the PM xerogels in different solvent systems. (a) PM/MeCN-H_2O (4% w/v, bar=50 μm); (b) PM/MeCN-H_2O (4% w/v, bar=10 μm); (c) PM/DMF-H_2O (4% w/v, bar=50 μm); (d) PM/DMF-H_2O (4% w/v, bar=10 μm)

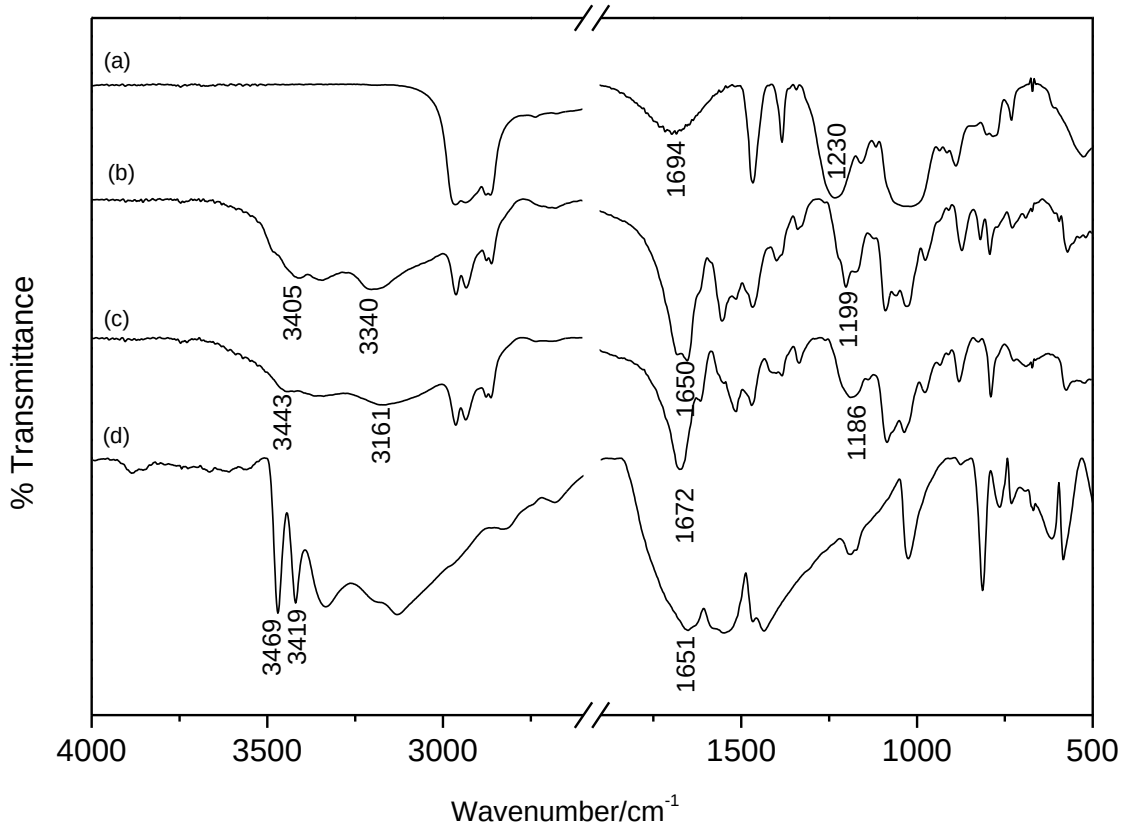


Figure S8. FTIR spectra of (a) pure P; (b) xerogel of PM/EG-H_2O (4% w/v); (c) xerogel of PM/DMF-H_2O(4% w/v); (d) pure M.

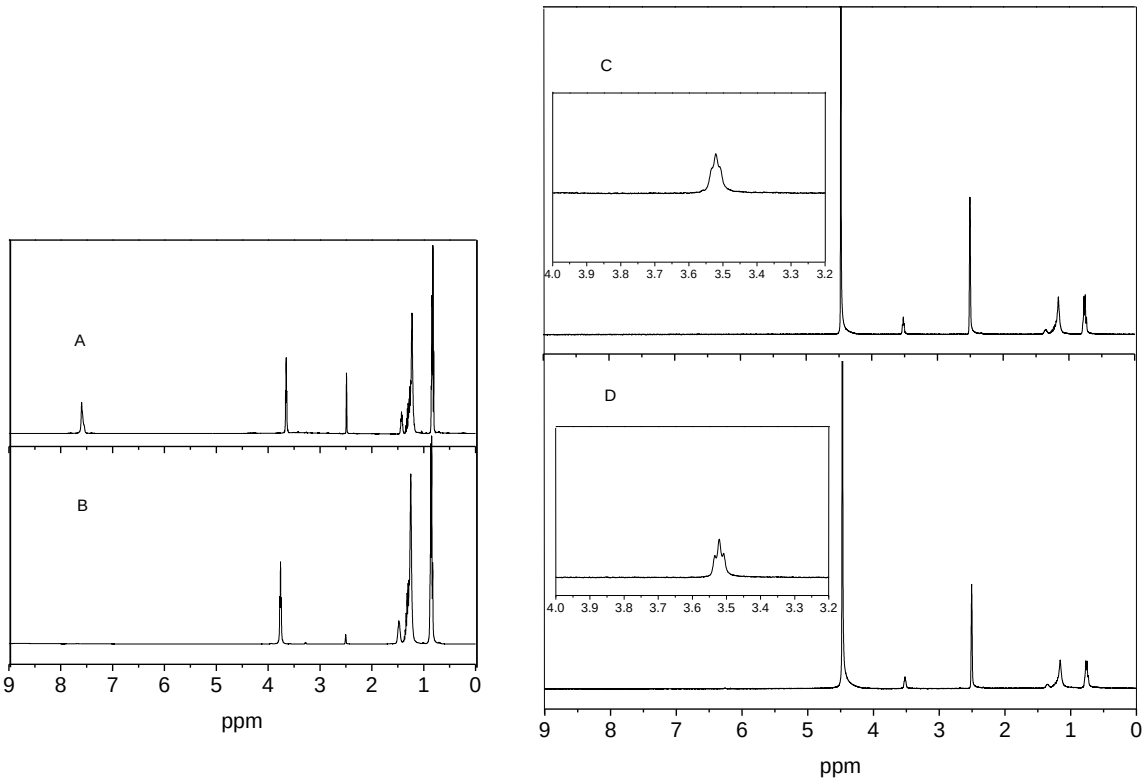


Figure S9. ^{1}H NMR spectra of PM in DMSO-d_6 (A), pure P in DMSO-d_6 (B), the mixed-hydrogel system of their sol (C) and gel (D) in D_2O-DMSO-d_6 (V/V=1:1). The insets show an expanded portion.

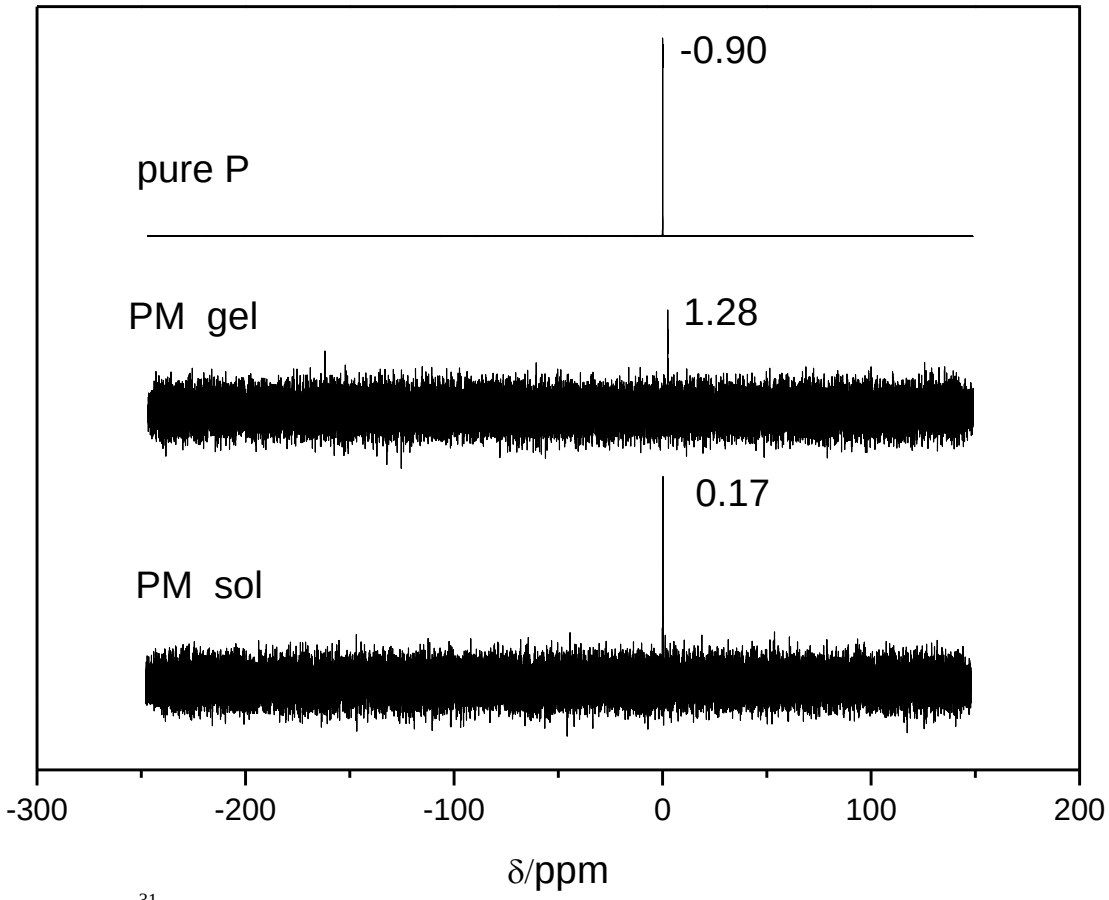


Figure S10. ^{31}P-NMR spectra of pure P, the mixed-hydrogel system of their sol and gel state in D_2O-DMSO-d_6 (V/V=1:1).

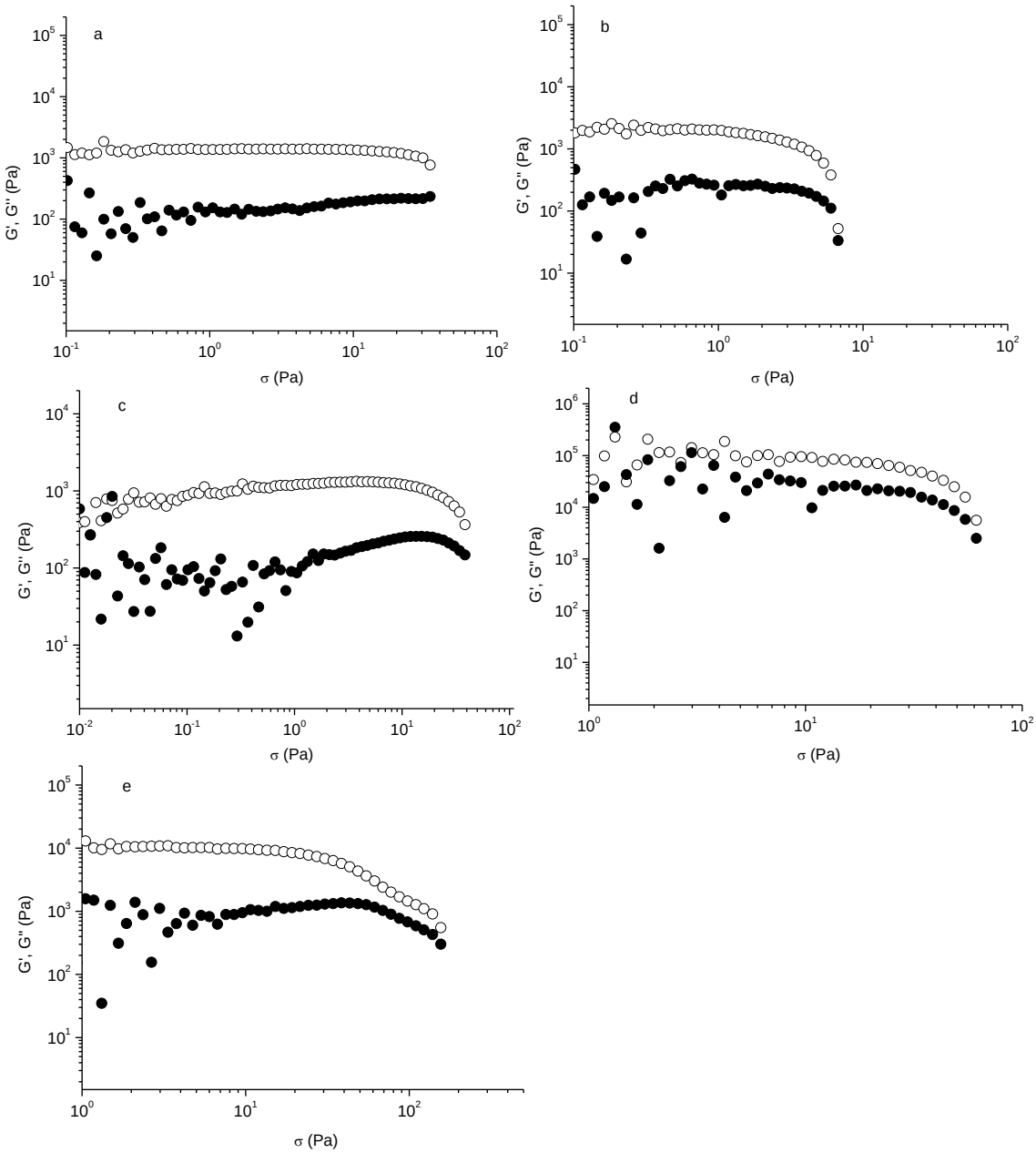


Figure S11. Measurement of the evolution of G′ and G″ at a frequency of 1 Hz and a temperature of 20 ℃, as a function of oscillation stress (σ) of the gels (4%, w/v) in (a) H_2O, (b) DMF-H_2O (1/1), (c) MeCN-H_2O (1/1), (d) EtOH-H_2O (1/1), (e) EG-H_2O (1/1): G′, open circles; G″, closed circles.

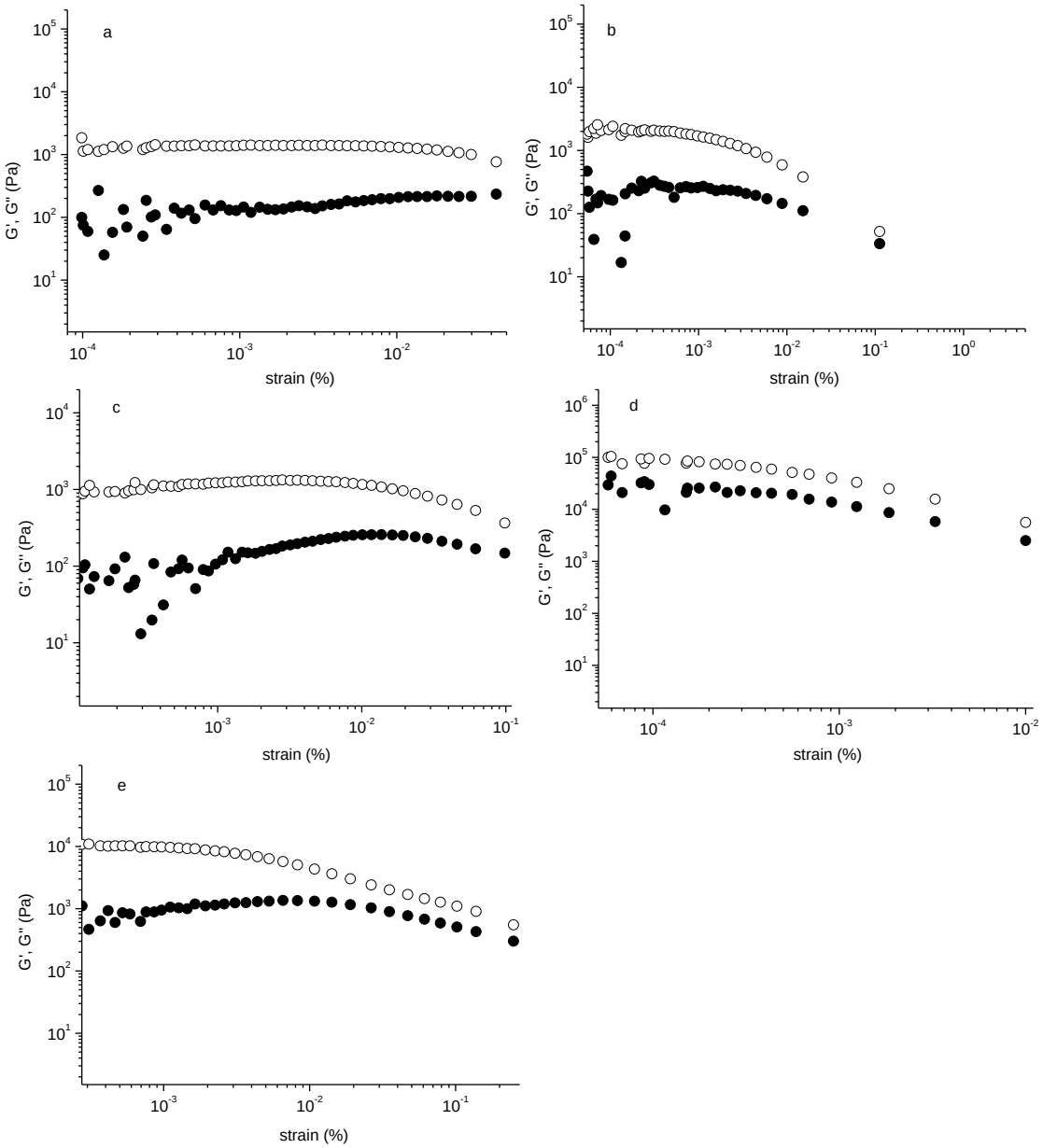


Figure S12. Log-log strain sweep (1.0 rad/sec) for the gels (4%, w/v) in (a) H_2O, (b) DMF-H_2O (1/1), (c) MeCN-H_2O (1/1), (d) EtOH-H_2O (1/1), (e) EG-H_2O (1/1): G′, open circles; G″, closed circles.

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