

# Amyloid Peptide Mixtures: Self-Assembly, Hydrogelation, Nematic Ordering and Catalysts in Aldol Reactions

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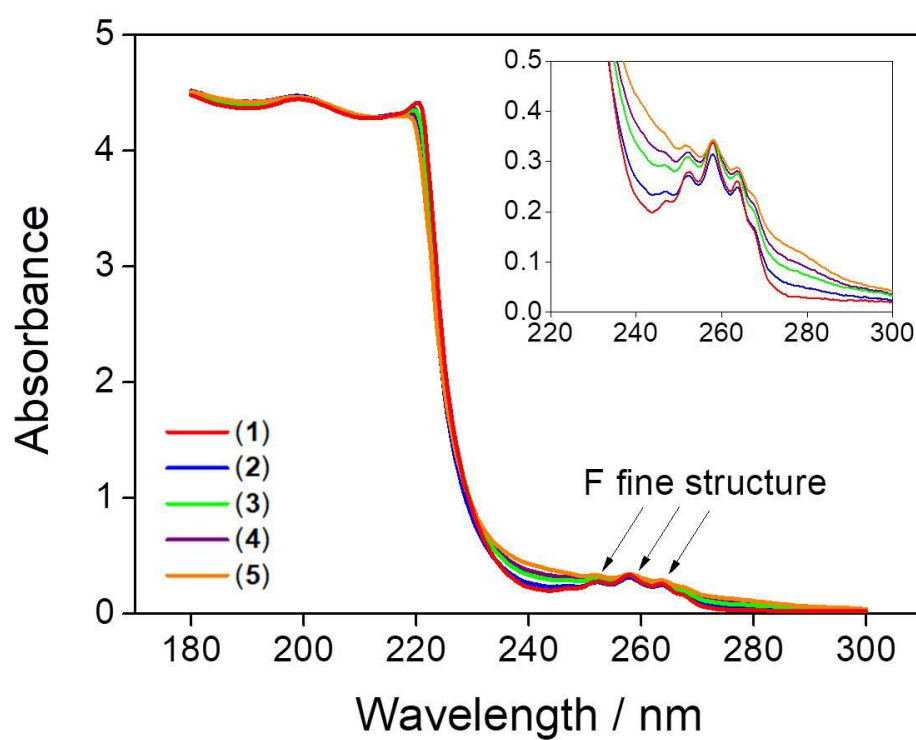
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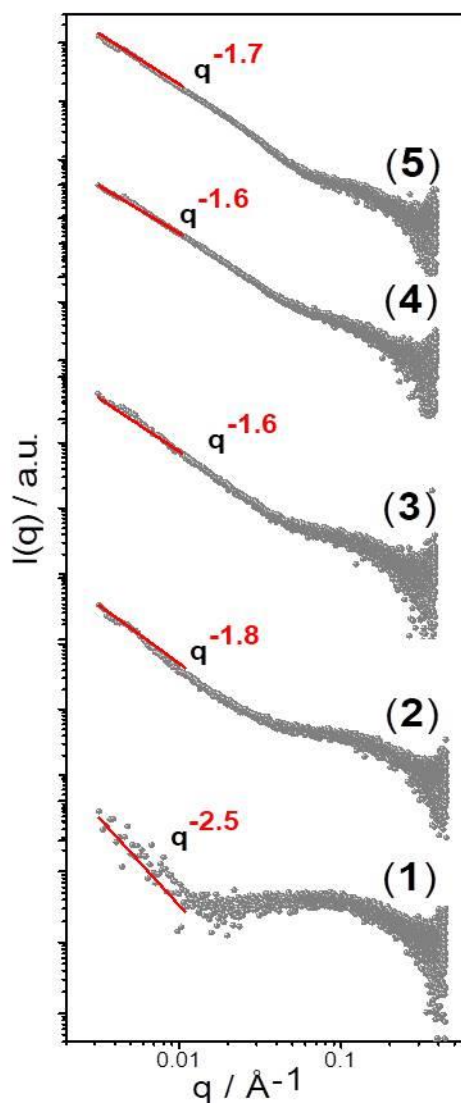
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**Table S1.** FTIR peak positions obtained using 0.5 wt% of P[RF]<sub>4</sub>:[RF]<sub>4</sub> mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**), at native pH.

Sample	Vibration (cm <sup>-1</sup> )						
	1	2	3	4	5	6	7
(1)	1672	1641	1607	1585	1457	1435	1368
(2)	1673	1643	1609	1585	1457	1438	1365
(3)	1673	1638	1607	1585	1450	1433	1368
(4)	1673	1646	1610	1585	1455	1438	1366
(5)	1672	1639	1609	1586	1459	1438	-

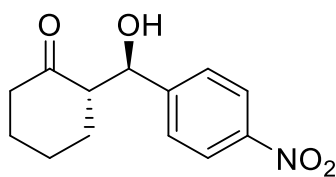


**Figure S1.** Absorption spectra of P[RF]<sub>4</sub>:[RF]<sub>4</sub> mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**), above the *cac* in water. Insert: amplified bands of the phenylalanine peaks.

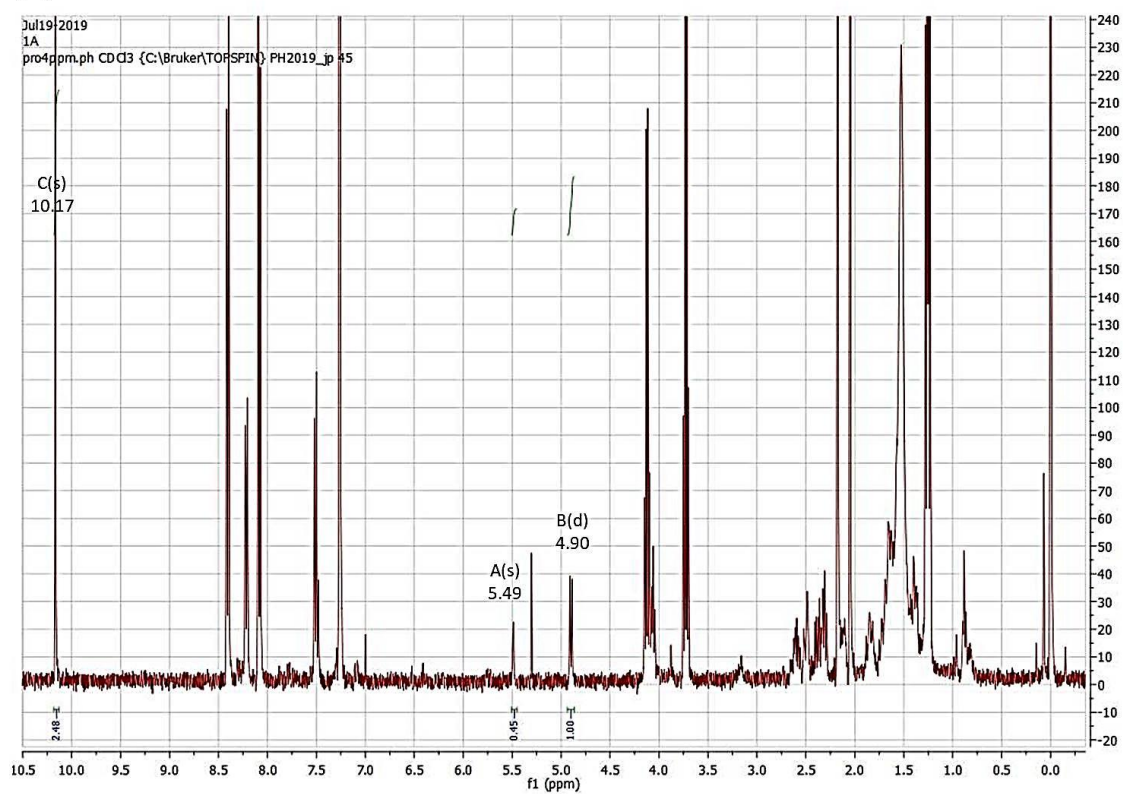


**Figure S2.** SAXS data showing linear fit in the Guinier regime for 0.5 wt% of of P[RF]<sub>4</sub>:[RF]<sub>4</sub> mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**).

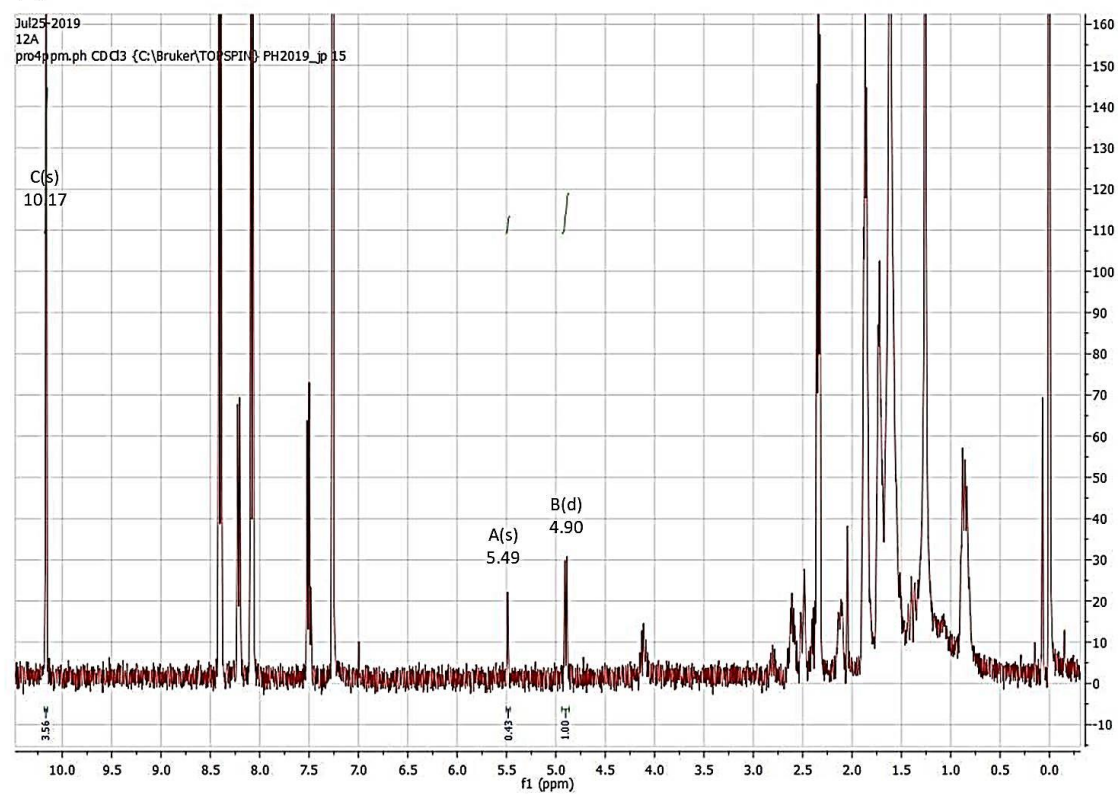
**(*S*)-2-((*R*)-Hydroxy(4-nitrophenyl)methyl)cyclohexan-1-one**



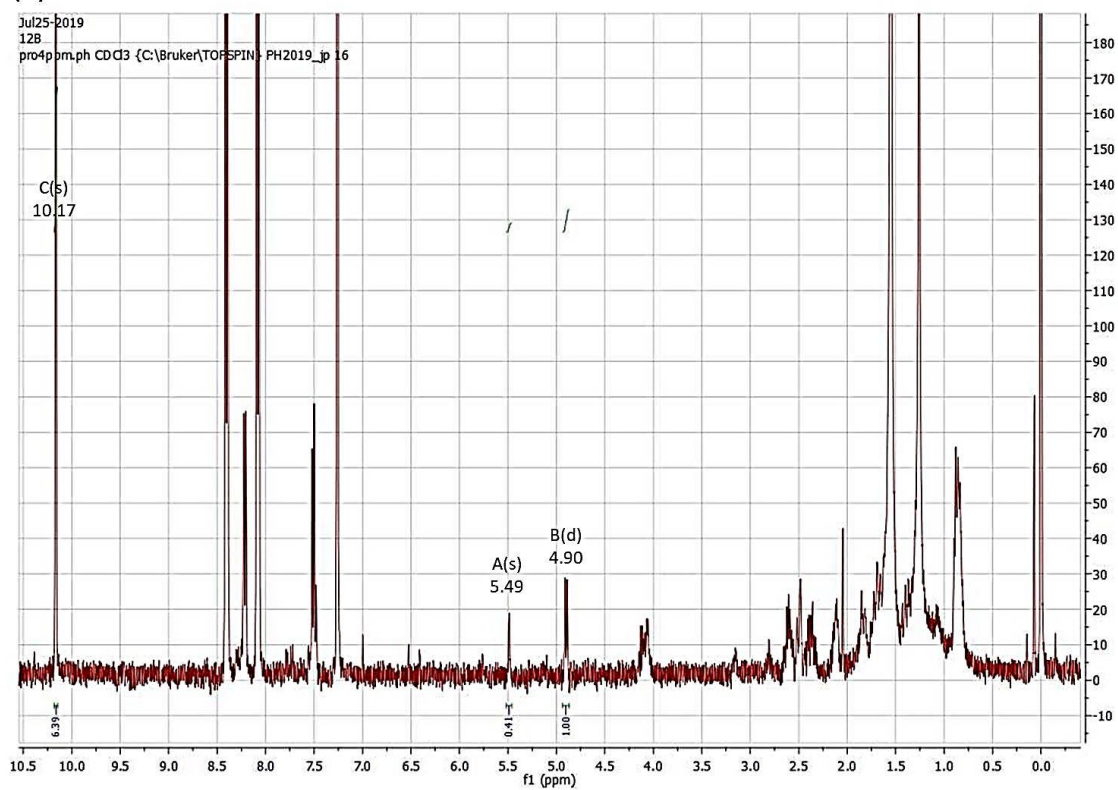
(1) 5 mol%



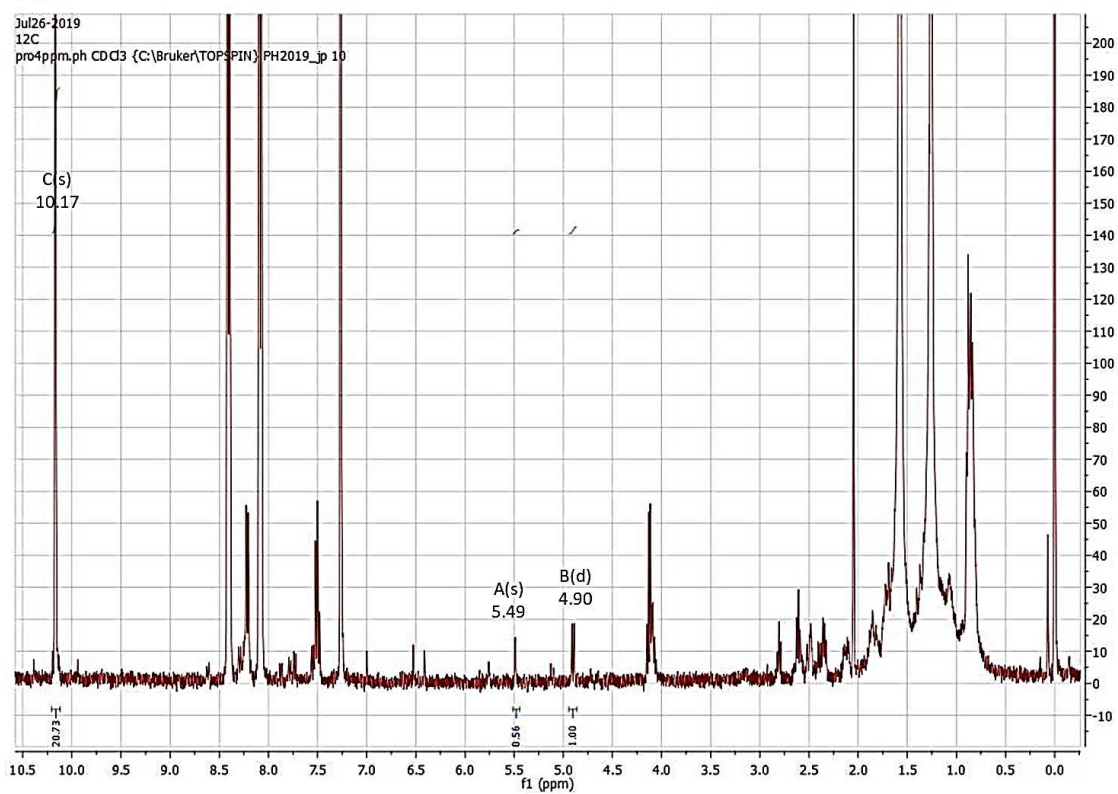
(2) 5 mol%



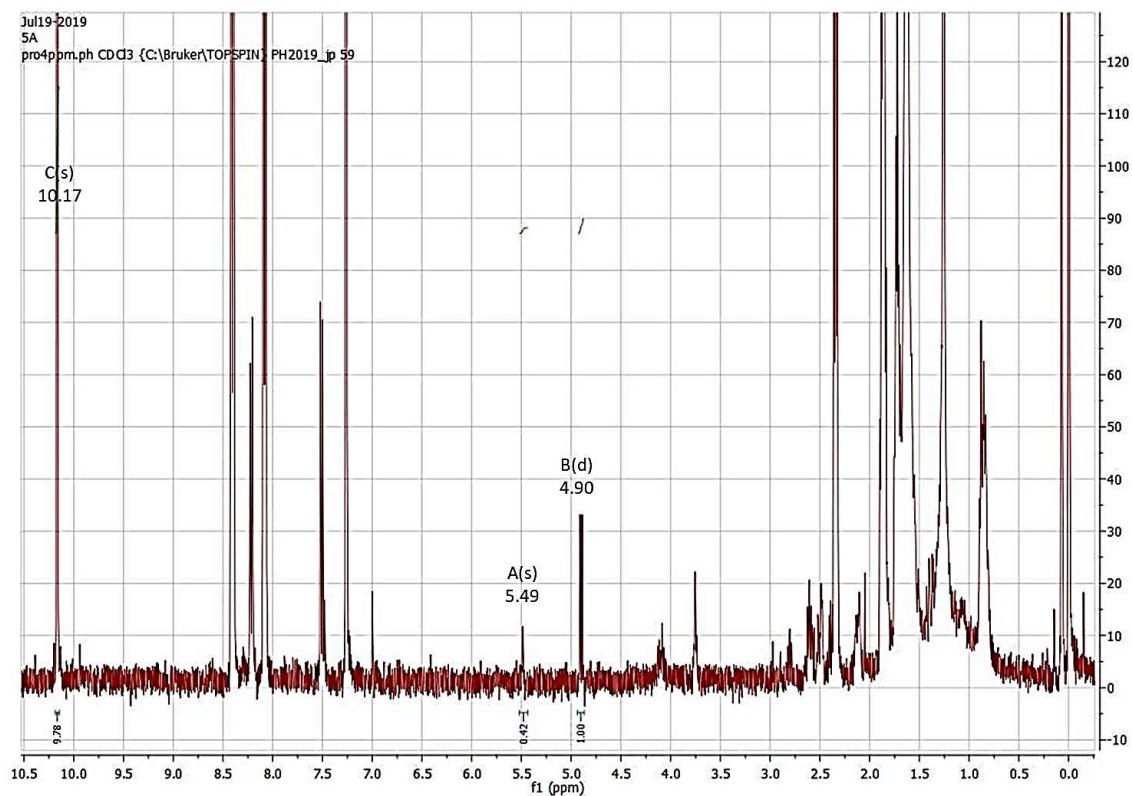
### (3) 5 mol%



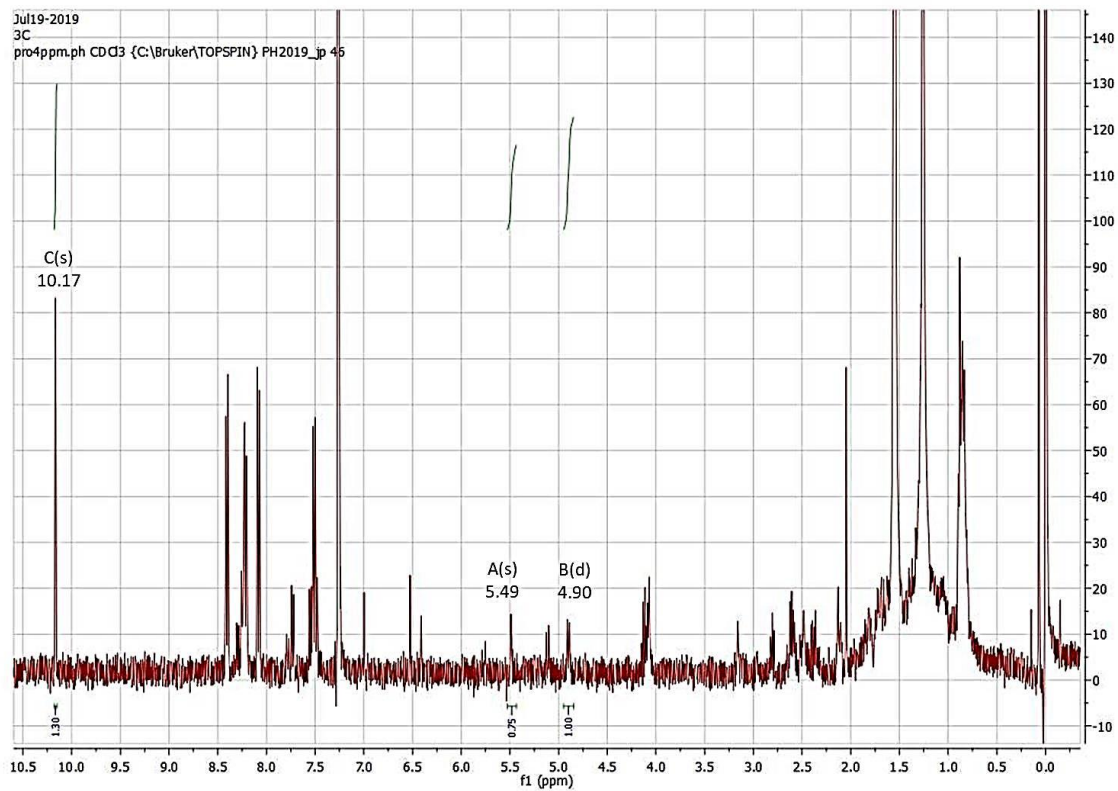
### (4) 5 mol%



(5) 5 mol%

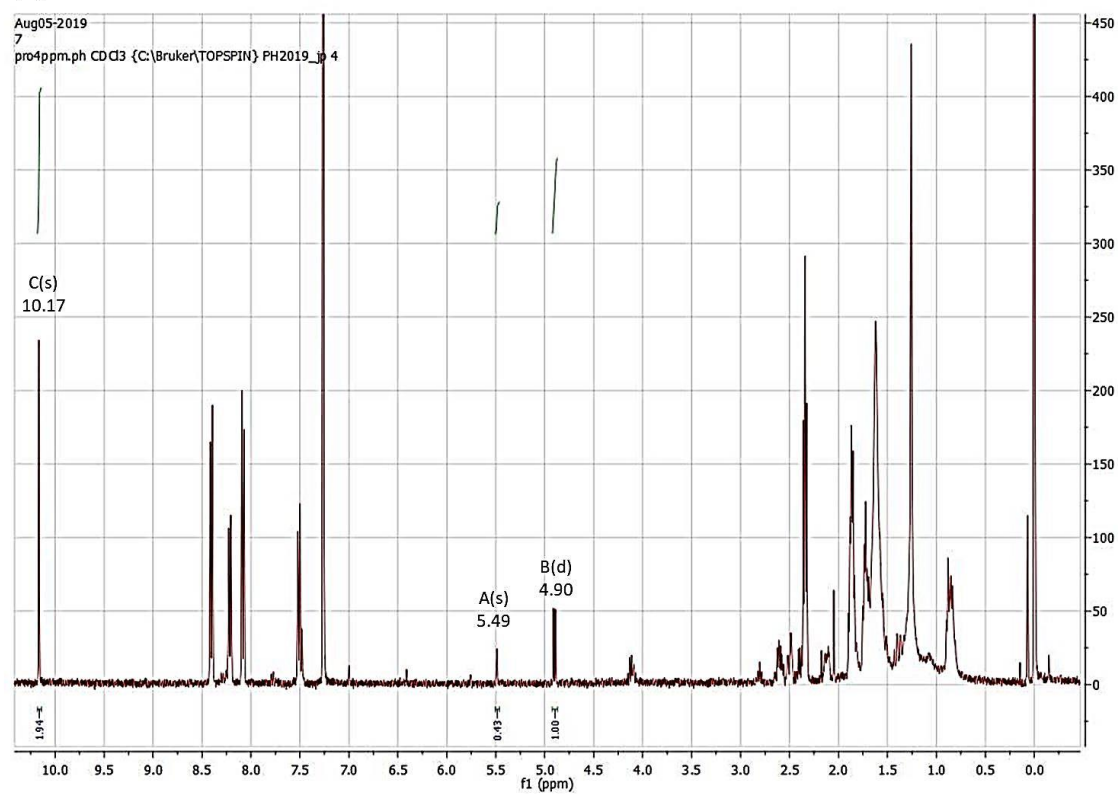


(1) 20 mol%

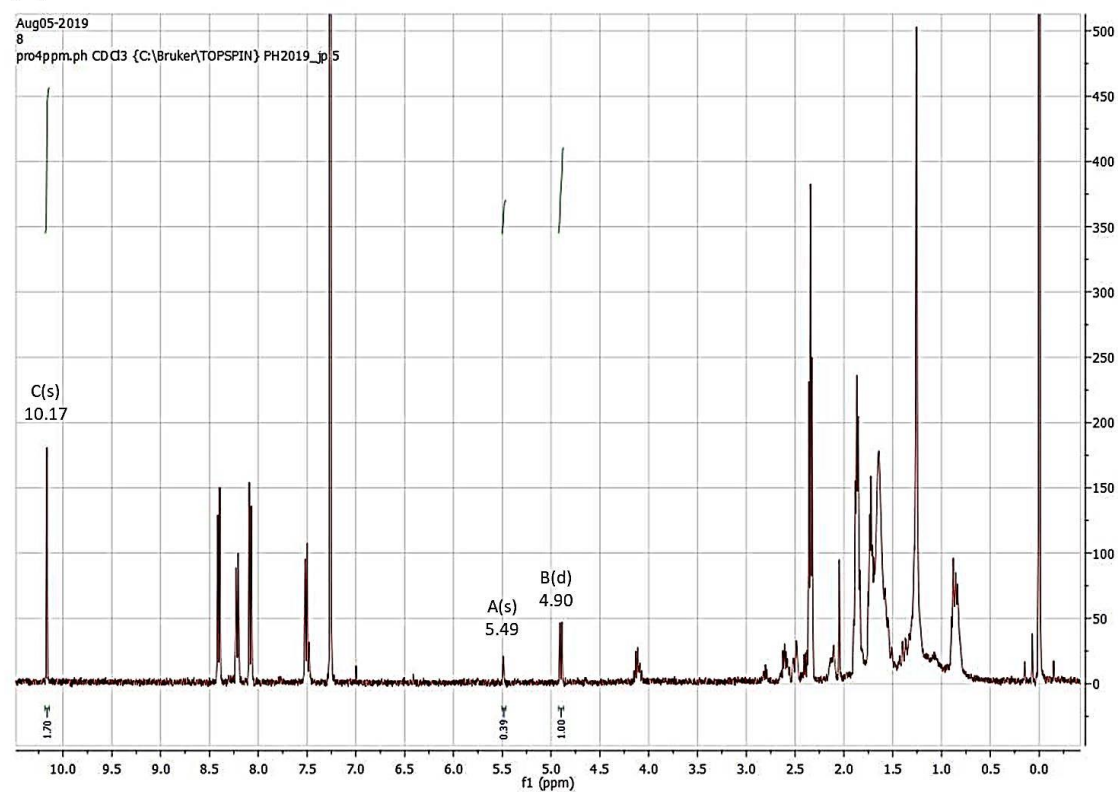




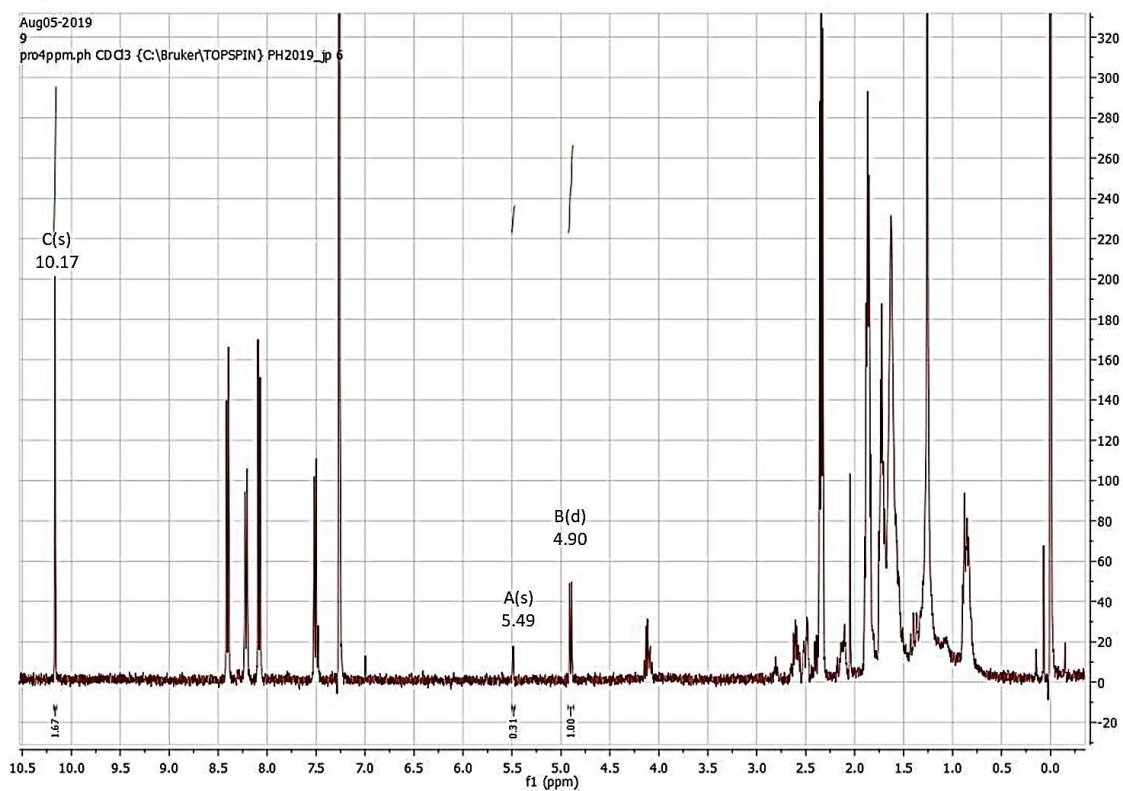
## (2) 20 mol%



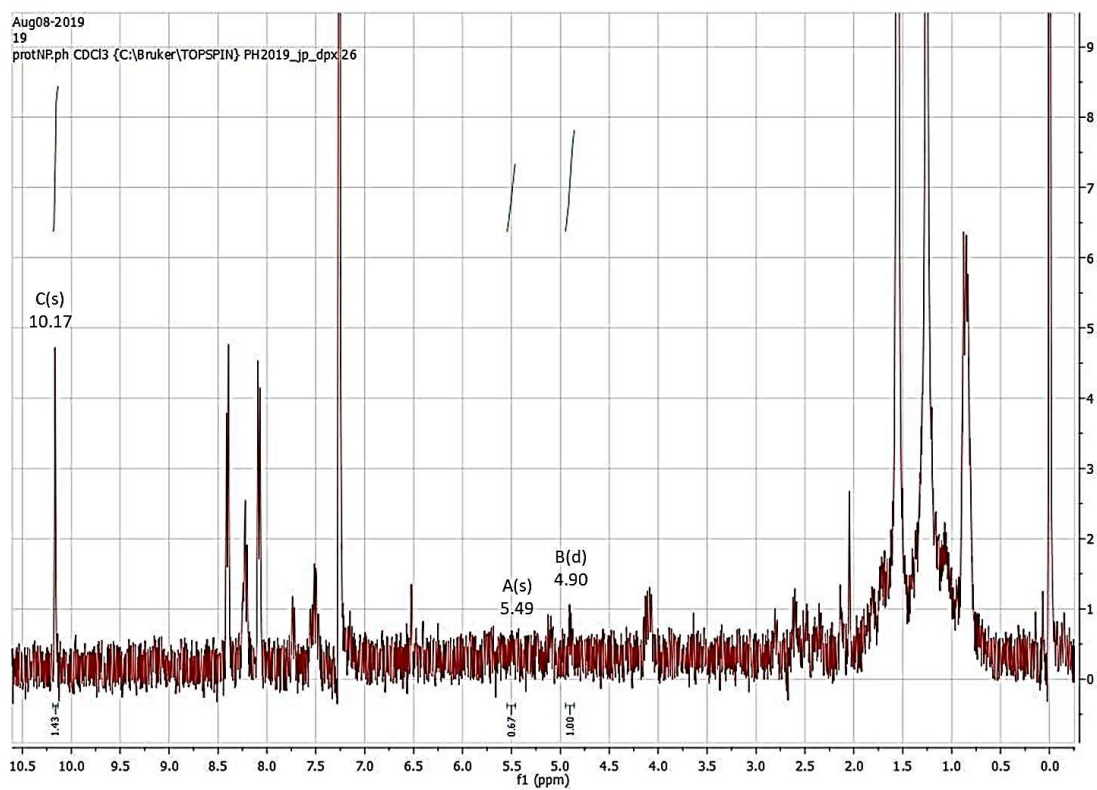
## (3) 20 mol%



(4) 20 mol%



(5) 20 mol%



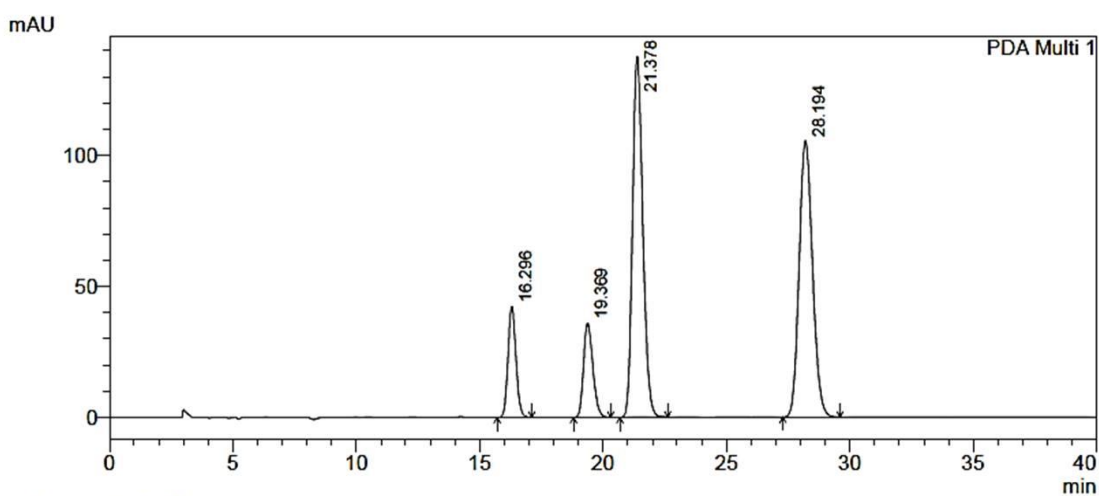
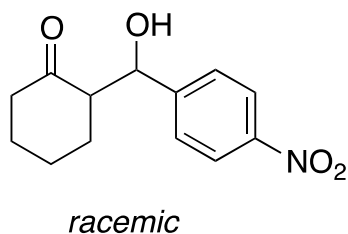
**$^1\text{H}$  NMR** (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.22-8.18 (m, 2H, ArH), 7.51-7.47 (m, 2H, ArH), 5.49 (br s, 1H, CHOH of *syn* diastereoisomer), 4.90 (dd,  $J = 7.5$  Hz, 3.0 Hz, 1H, CHOH of *anti*



diastereoisomer), 2.66-2.30 (m, 1H,  $\text{CHCHOH}$ ), 2.66-2.30 (m, 2H,  $\text{CH}_2\text{C(O)}$ ), 2.16-1.24 (m, 6H, chex-H).

**Figure S3.** Representative  $^1\text{H}$  NMR spectra of crude aldol products of  $\text{P[RF]}_4\text{: [RF]}_4$  mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**), considering 5 mol% and 20 mol% of catalyst, described in Table 3.

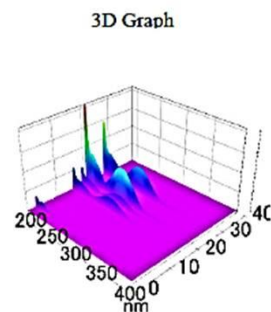
### Chiral-phase HPLC analysis



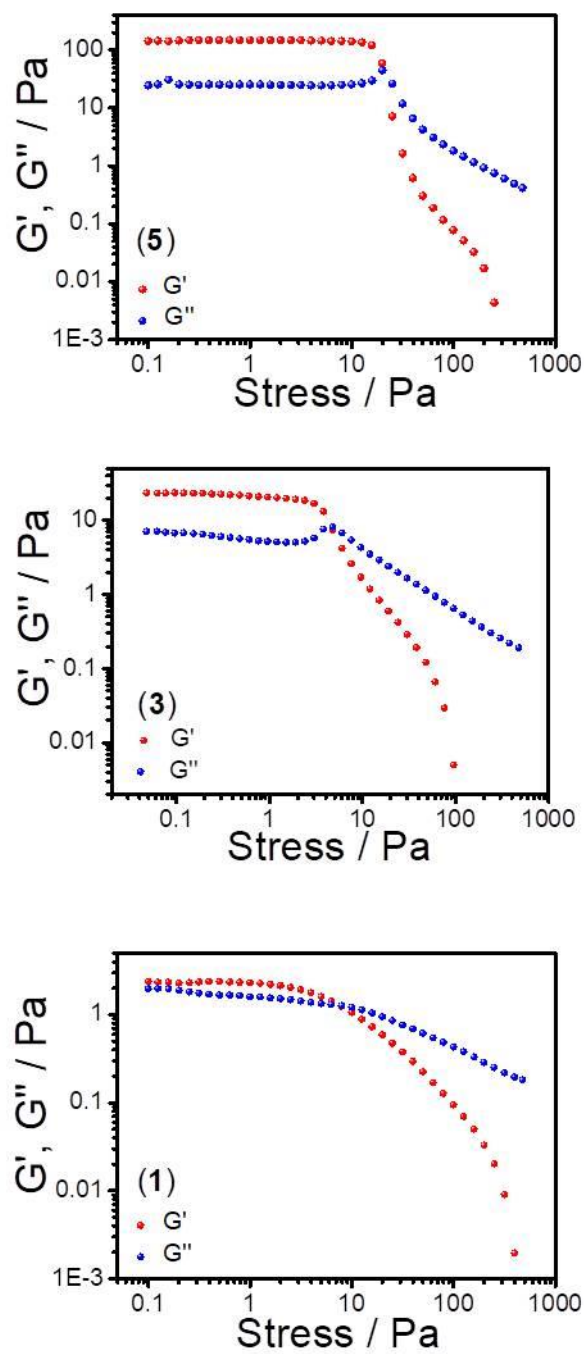
1 PDA Multi 1/254nm 4nm

PeakTable

PDA Ch1 254nm 4nm					
Peak#	Ret. Time	Area	Height	Area %	Height %
1	16.296	943300	42301	9.594	13.158
2	19.369	939071	35915	9.551	11.171
3	21.378	3974790	137730	40.428	42.841
4	28.194	3974557	105542	40.426	32.829
Total		9831718	321489	100.000	100.000



**Figure S4.** HPLC chromatogram for racemic aldol product. Conditions: Chiralpak AD-H, hexane/2-propanol (90/10);  $1.0 \text{ mL} \cdot \text{min}^{-1}$ ,  $\lambda = 254 \text{ nm}$ .



**Figure S5.** Storage and shear moduli obtained by rheology experiments in stress sweep mode of mixtures (1), (3), and (5).

**Table S2.** Fit parameters from SAXS for 3 wt% hydrogels of mixtures **(1)**, **(3)**, and **(5)**.

Hydrogel	Long Cylindrical Shell				
	<b>R</b> (nm)	<b><math>\sigma_R</math></b> (nm)	<b><math>\Delta R</math></b> (nm)	<b><math>\eta_{\text{core}}</math></b>	<b><math>\eta_{\text{shell}}</math></b>
<b>(1)</b>	2.5	0.18	0.68	$1.6 \times 10^{-7}$	$9.1 \times 10^{-9}$
<b>(3)</b>	2.3	0.45	0.50	$2.1 \times 10^{-7}$	$1.1 \times 10^{-7}$
<b>(5)</b>	0.7	0.07	0.61	$-7.4 \times 10^{-7}$	$3.5 \times 10^{-6}$