

**Supporting Information**

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

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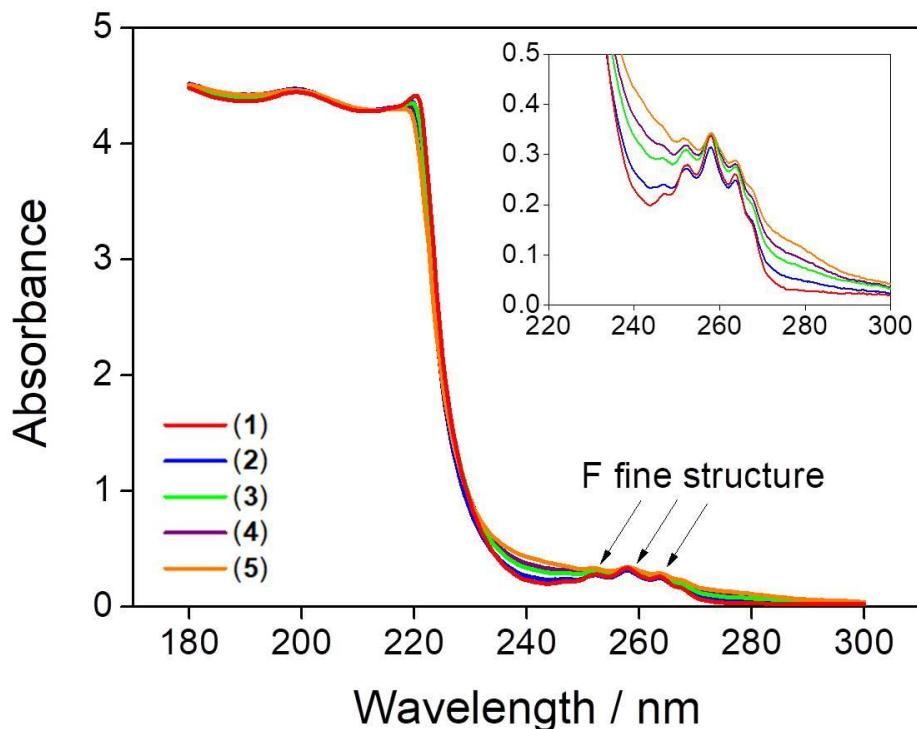
Number of figures: 5

Number of tables: 2

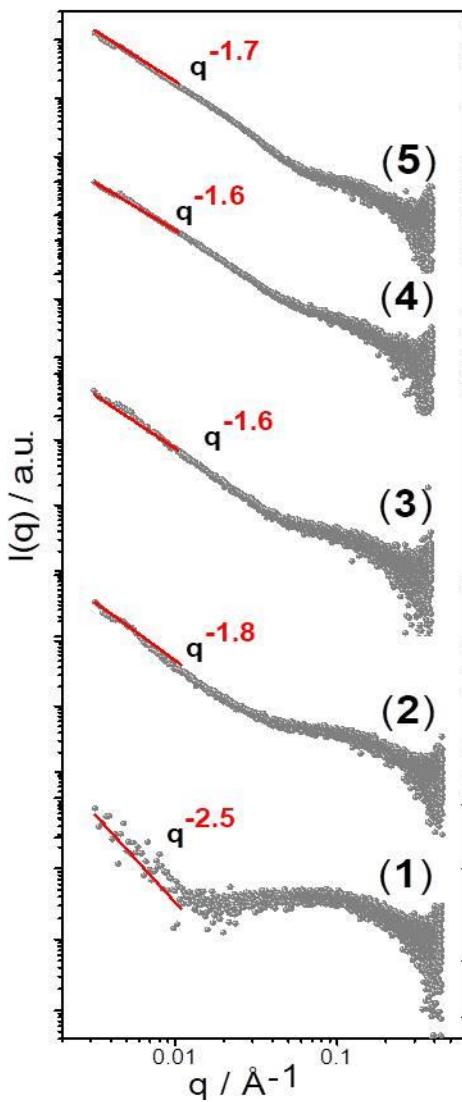
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**Table S1.** FTIR peak positions obtained using 0.5 wt% of P[RF]<sub>4</sub>:[RF]<sub>4</sub> mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**), at native pH.

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

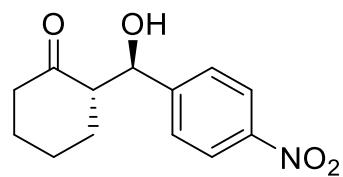


**Figure S1.** Absorption spectra of P[RF]<sub>4</sub>:[RF]<sub>4</sub> mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**), above the *cac* in water. Insert: amplified bands of the phenylalanine peaks.

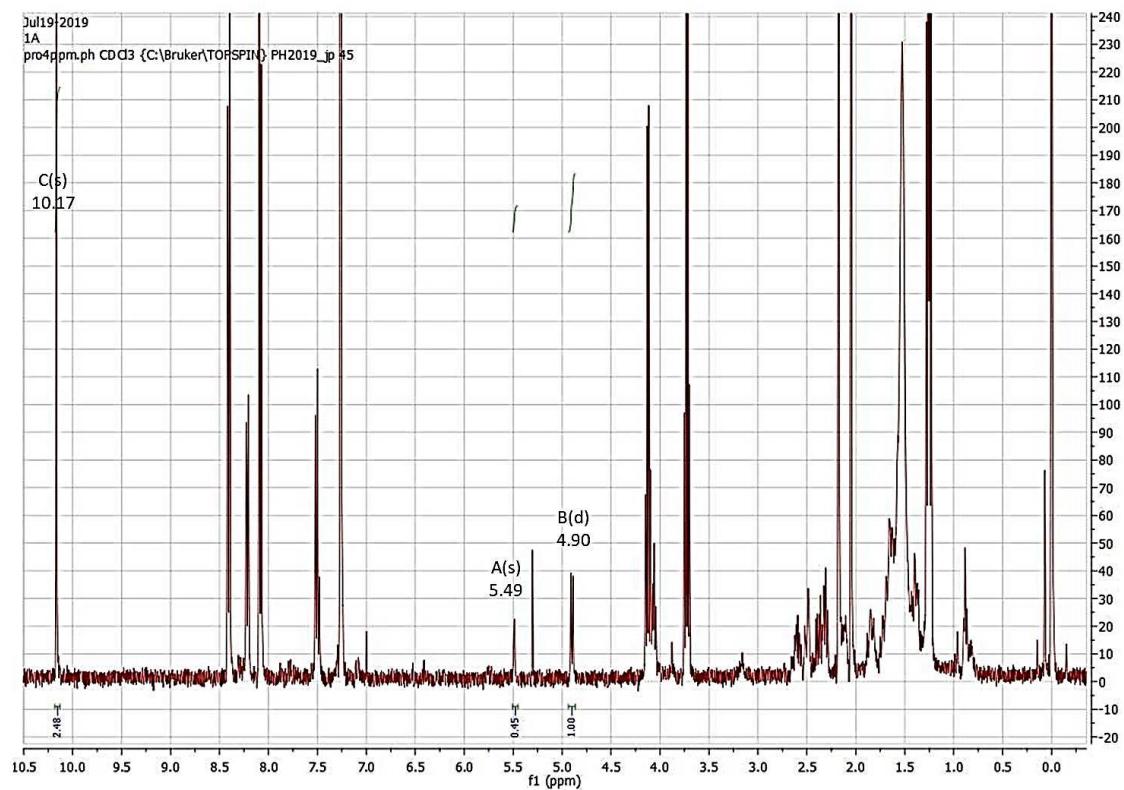


**Figure S2.** SAXS data showing linear fit in the Guinier regime for 0.5 wt% of P[RF]<sub>4</sub>:[RF]<sub>4</sub> mixtures 0:1 (**1**), 3:7 (**2**); 5:5 (**3**), 7:3 (**4**), and 1:0 (**5**).

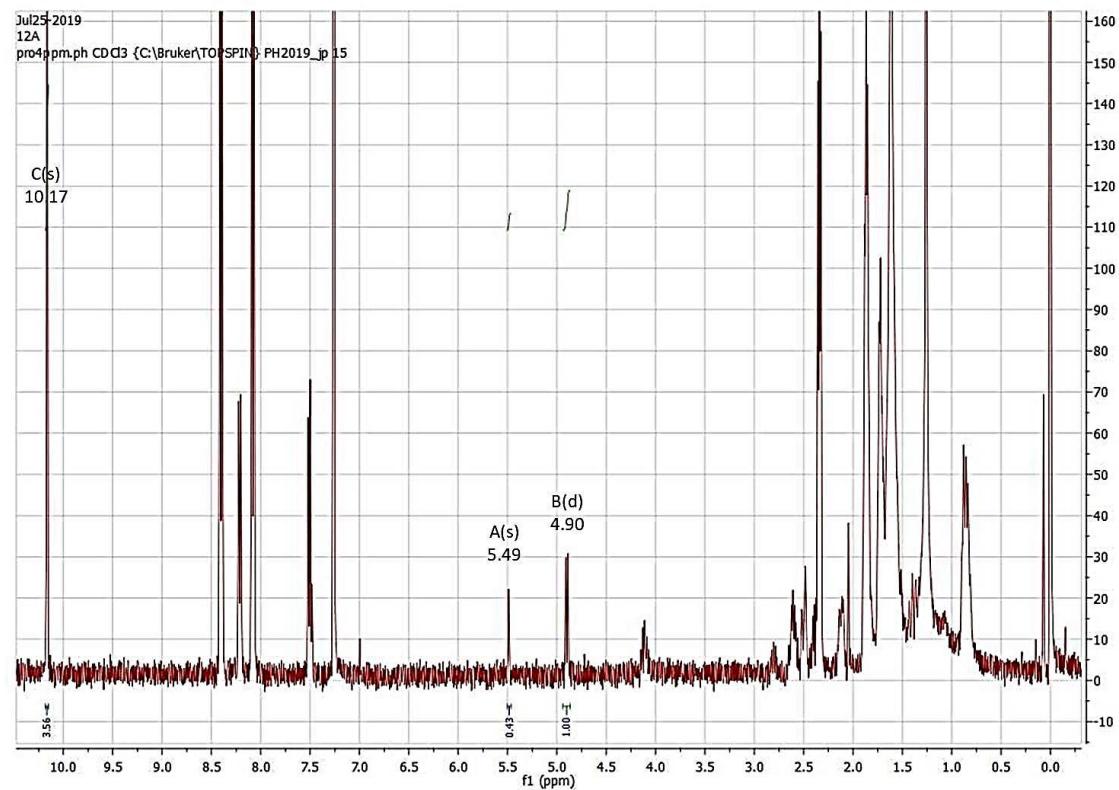
### (*S*)-2-((*R*)-Hydroxy(4-nitrophenyl)methyl)cyclohexan-1-one



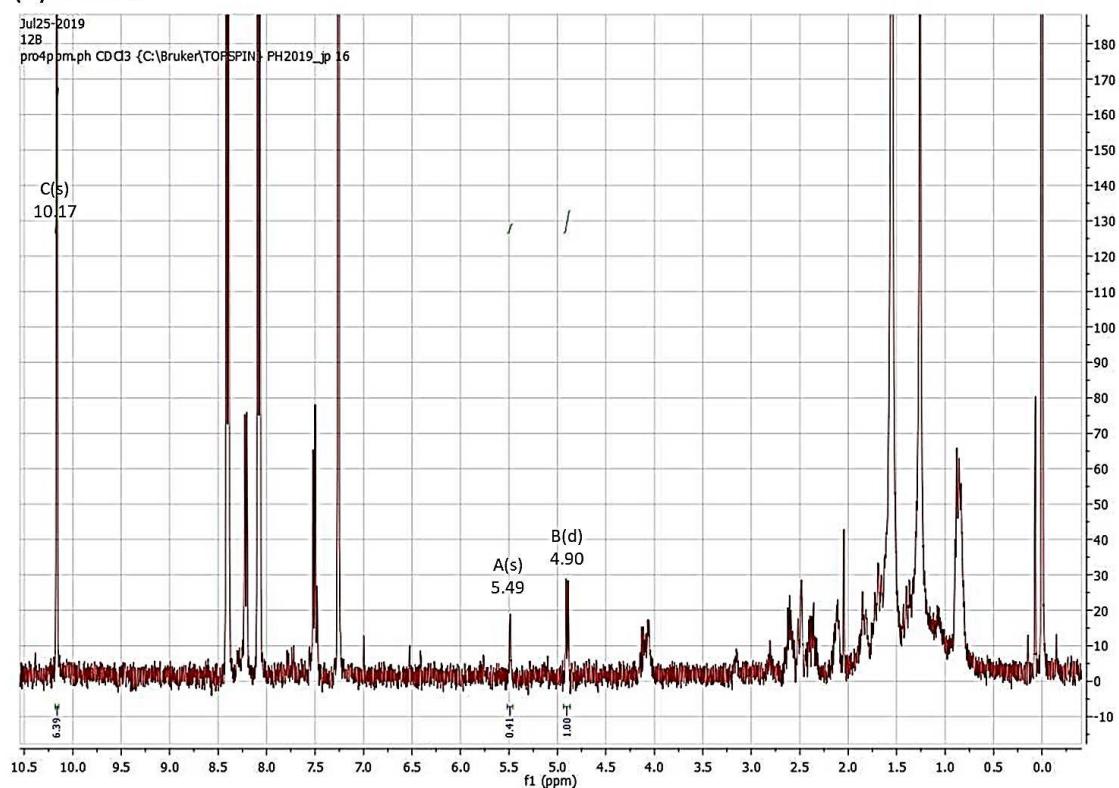
**(1) 5 mol%**



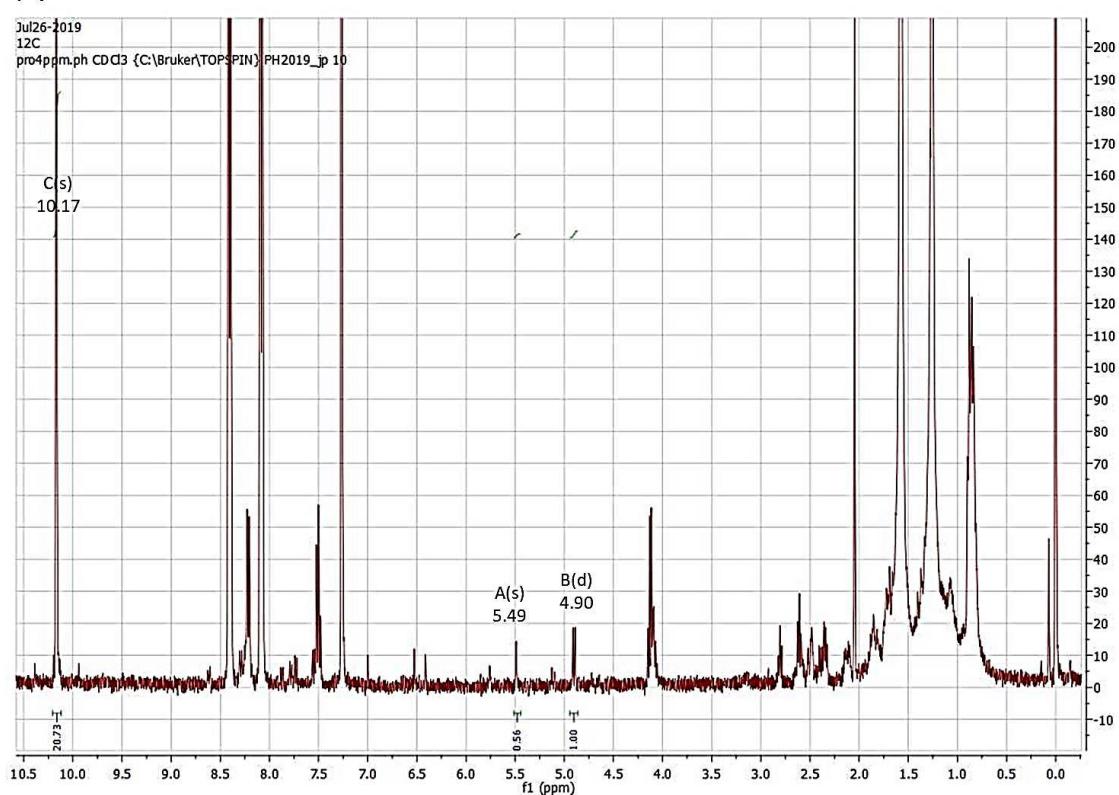
**(2) 5 mol%**



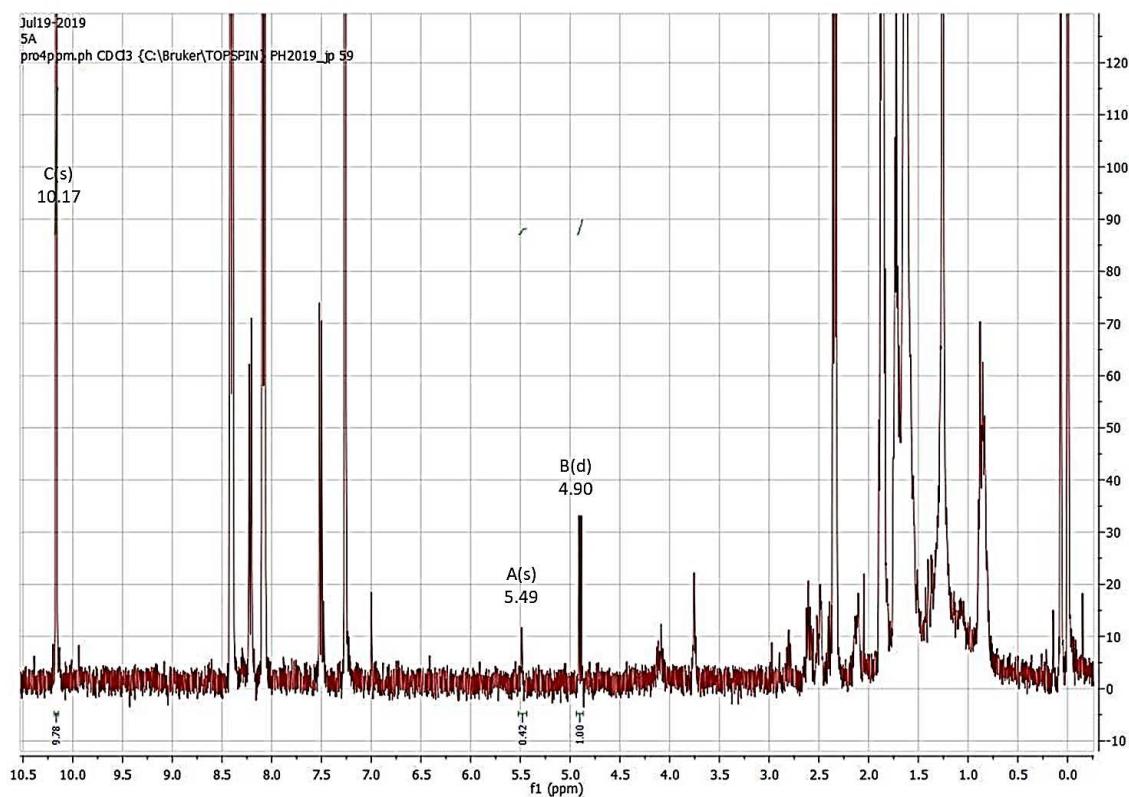
**(3) 5 mol%**



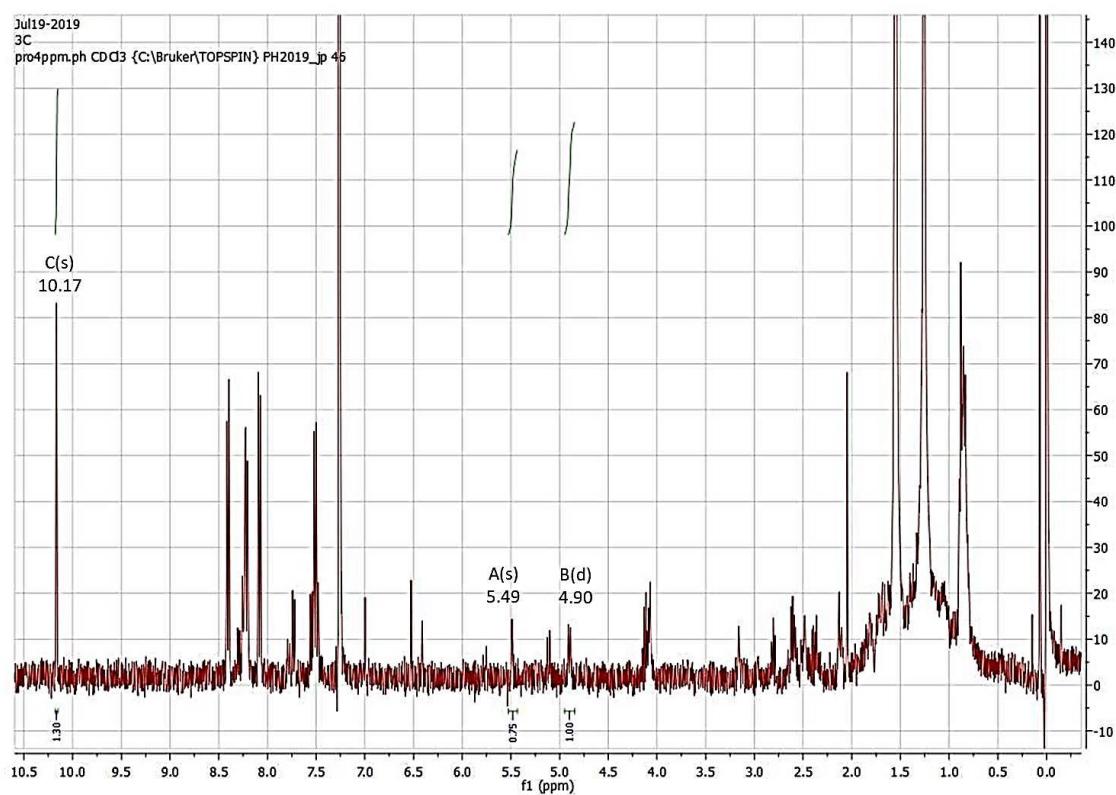
**(4) 5 mol%**



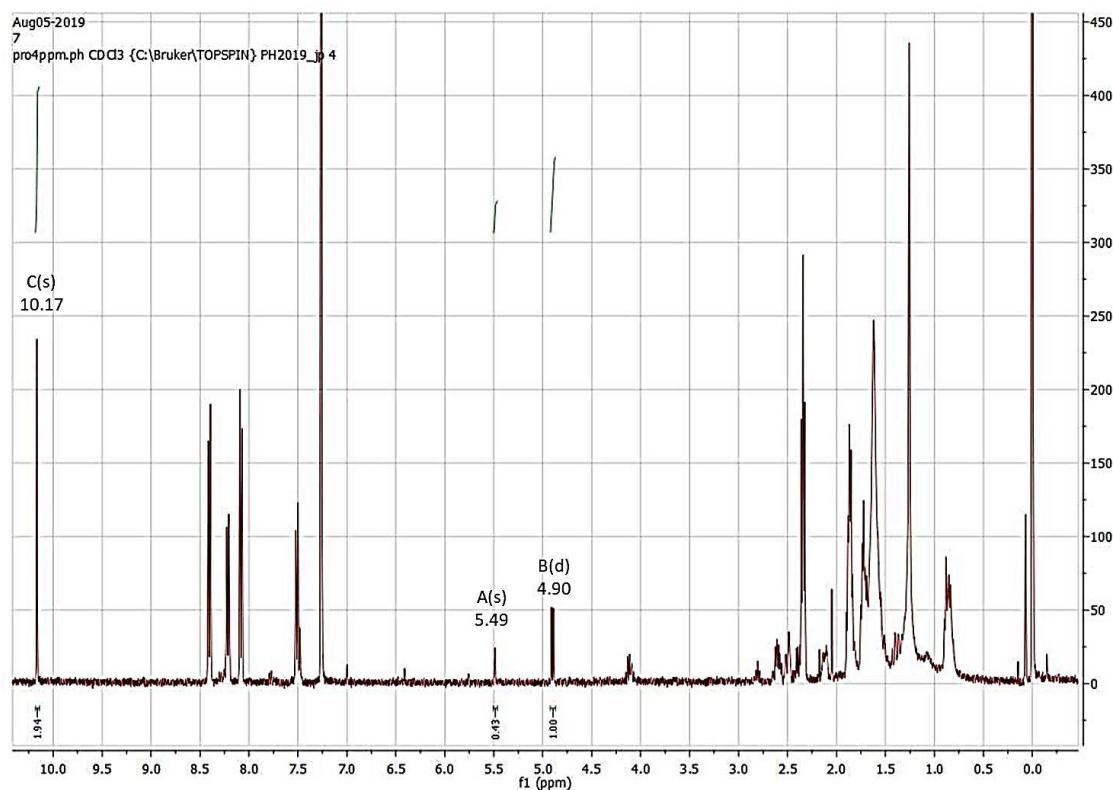
**(5) 5 mol%**



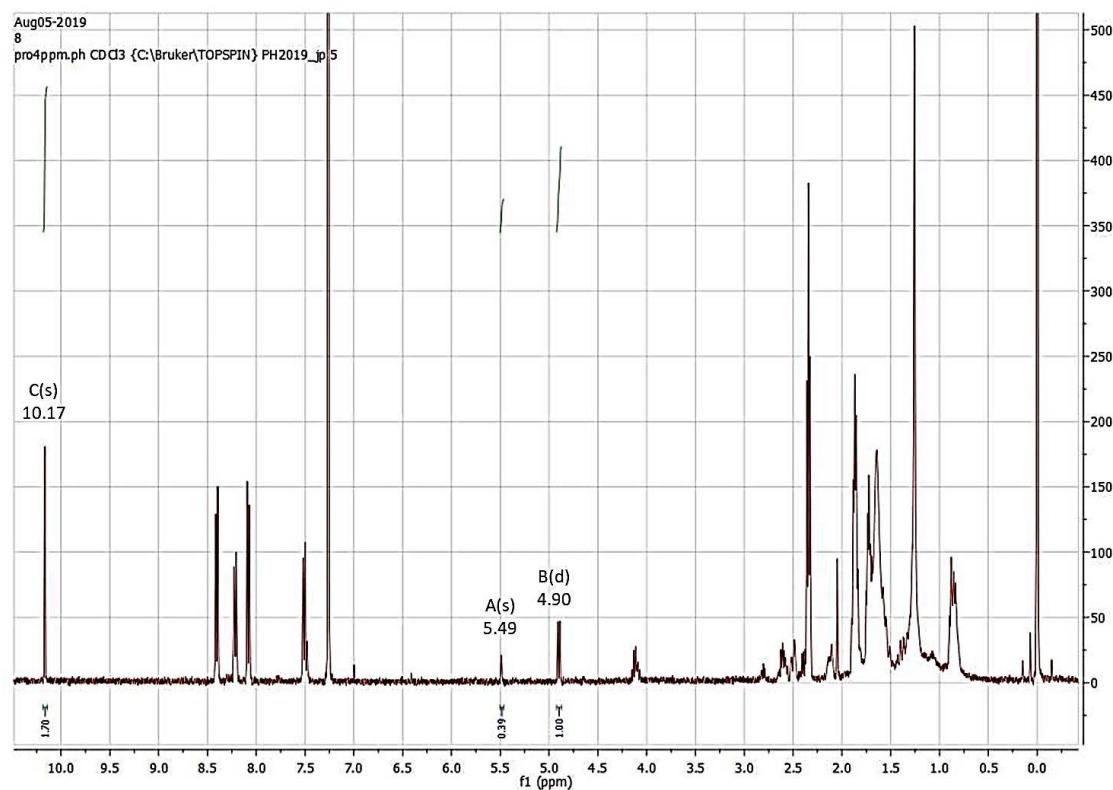
**(1) 20 mol%**



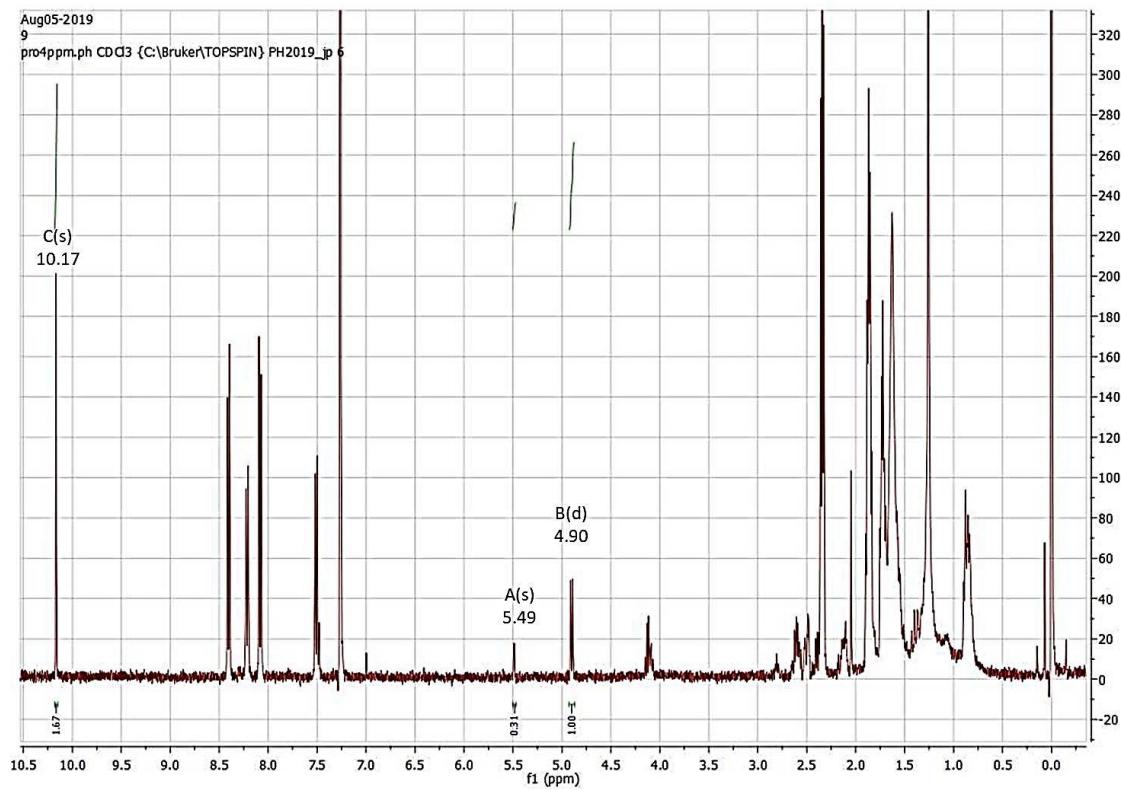
(2) 20 mol%



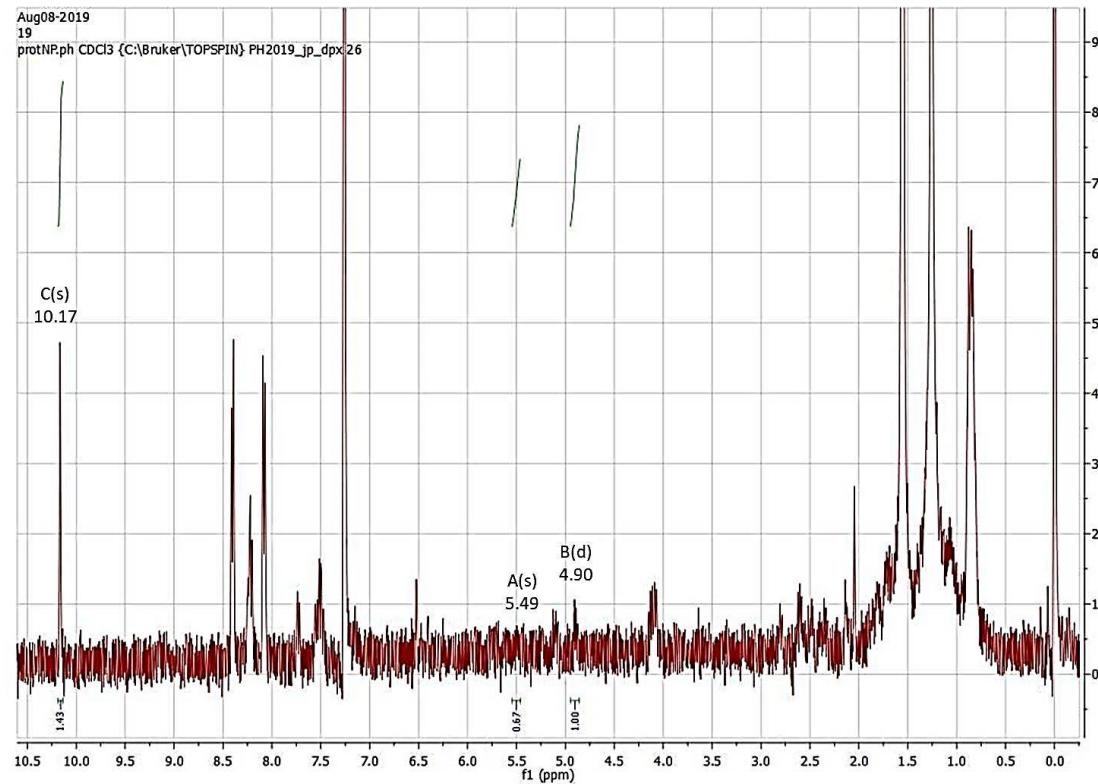
(3) 20 mol%



**(4) 20 mol%**



**(5) 20 mol%**

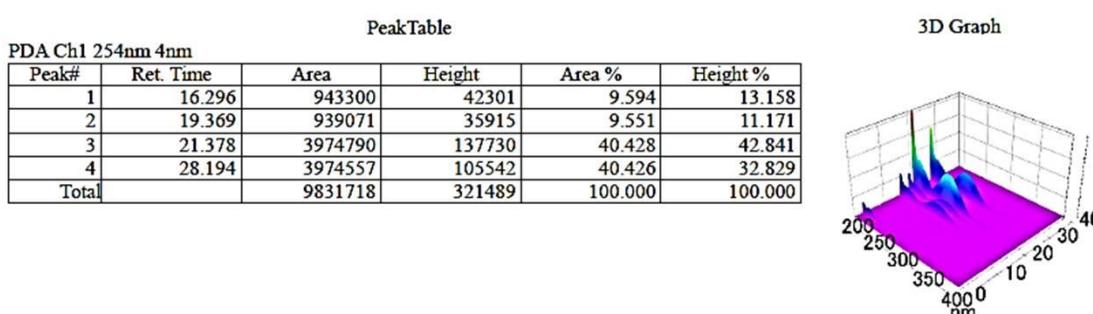
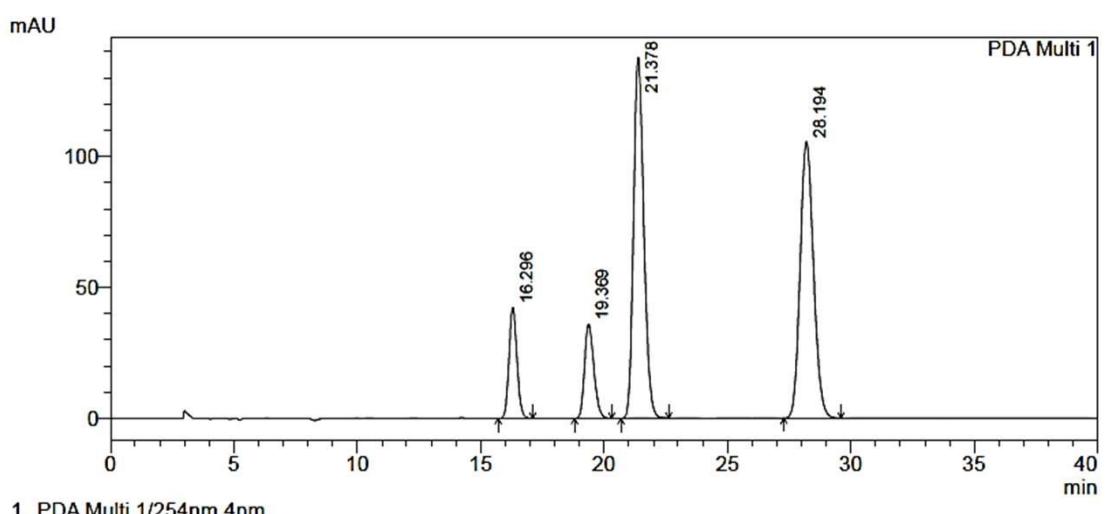
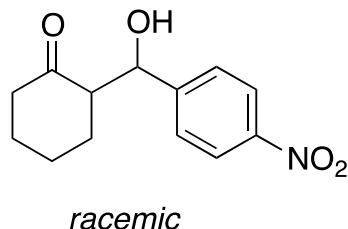


**<sup>1</sup>H NMR** (300 MHz, CDCl<sub>3</sub>): δ 8.22-8.18 (m, 2H, ArH), 7.51-7.47 (m, 2H, ArH), 5.49 (br s, 1H, CHOH of *syn* diastereoisomer), 4.90 (dd, J = 7.5 Hz, 3.0 Hz, 1H, CHOH of *anti*

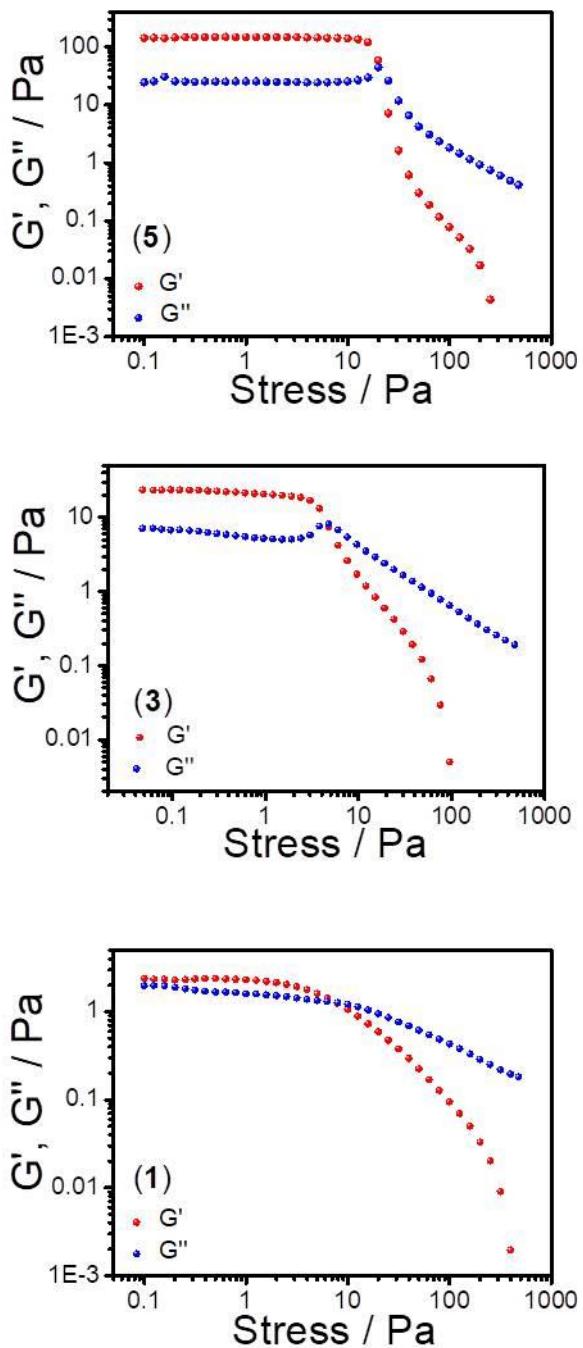
diastereoisomer), 2.66-2.30 (m, 1H, CHCHOH), 2.66-2.30 (m, 2H, CH<sub>2</sub>C(O)), 2.16-1.24 (m, 6H, chex-H).

**Figure S3.** Representative <sup>1</sup>H NMR spectra of crude aldol products 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**), considering 5 mol% and 20 mol% of catalyst, described in Table 3.

### Chiral-phase HPLC analysis



**Figure S4.** HPLC chromatogram for racemic aldol product. Conditions: Chiraldak AD-H, hexane/2-propanol (90/10); 1.0 mL·min<sup>-1</sup>,  $\lambda=254$  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				
	R (nm)	$\sigma_R$ (nm)	$\Delta R$ (nm)	$\eta_{core}$	$\eta_{shell}$
<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	$-7.4 \times 10^{-7}$	$3.5 \times 10^{-6}$