**Supporting Information** 

## Novel Supramolecular Polymer Fabricated via Stronger Hydrogen-Bonding Interactions between Substituted Amide Groups: Design, Synthesis, Properties and Mechanism

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Synthesis of di(2-pyridine carbaldehyde)-hexanediohydrazide (DPCH) from adipic dihydrazide (ADH)

DPCH was synthesized through a one-step reaction process as indicated in Scheme S1.

The solution of 2-pyridinecarboxaldehyde (2) (6.42 g, 60.0 mmol) in ethanol (20 mL) was dropped into the suspension of adipic dihydrazide (ADH) (3.44 g, 20.0 mmol) with a drop of acetic acid in ethanol (50 mL) and the reaction was conducted at room temperature for 20 hrs. After filtration, the powder was washed by ethanol and water for two times to remove the unreacted 2-pyridinecarboxaldehyde and adipic dihydrazide. Then the white powder was dried in vacuum at 50 °C for 10 hours to remove any residual solvent and white powder of di(2-pyridine carbaldehyde)-hexanediohydrazide (DPCH) (5.04 g, in 72.0% yield) was obtained.





The structure of DPCH was confirmed <sup>1</sup>H-NMR. <sup>1</sup>H-NMR (DMSO-D6, 300 MHz, δ/ppm, Fig. S1): 1.63~2.67 (m, 8H, H atoms in the solid boxes, g), 7.01 (m, 2H, b), 7.87~7.99 (m, 4H, c and d), 8.15 (s, 2H, a), 8.45 (d, 2H, e), 11.46 (d, 2H, f). The disappearance of peaks at 4.1 ppm and 8.9 ppm also confirms that DPCH has been successfully prepared without ADH left. <sup>1</sup>H-NMR (DMSO-D6) of adipic dihydrazide (M<sub>2</sub>) was got from Reference S1.



Fig.S1 <sup>1</sup>H-NMR spectrum of DPCH.



Fig.S2 FT-IR spectrum of monomer DPCHP-DODE.



Fig.S3 (a) Shear test of lap joints for regular glasses bonded with DPCHP-DODE<sub>110</sub> and (b) cohesive failure for regular glasses bonded with DPCHP-DODE<sub>110</sub> after shear test.



Fig.S4 DSC curves of the (a) cooling process and (b) first heating process for monomer **DPCHP-DODE**.



Fig.S5 tan $\delta$  of DPCHP-DODE<sub>cooled</sub> and DPCH<sub>cooled</sub>.



Fig. S6 DSC curves for the first heating, cooling and second heating process of **DPCH**.



Fig.S7 FT-IR spectra of DPCHP-DODE from 40 to 180 °C ranged (a) from 3300 to

3100 cm<sup>-1</sup> and (b) from 1725 to 1625 cm<sup>-1</sup>, and **DPCHP-DODE**<sub>110</sub> from 40 to 180 ° C ranged (c) from 3300 to 3100 cm<sup>-1</sup> and (d) from 1725 to 1625 cm<sup>-1</sup>.



Fig.S8 FT-IR spectra of DPCHP-DODE and DPCHP-DODE<sub>110</sub> at 220 °C ranged (a) from 3300 to 2980 cm<sup>-1</sup> and (b) from 1725 to 1525 cm<sup>-1</sup>.



Fig.S9 TGA curves of (a) DPCHP-DODE and (b) DPCHP-DODE<sub>110</sub>.



**Fig.S10** FT-IR spectra of **DPCHP-DODE** in the cooling process ranged (a) from 3300 to 3170 cm<sup>-1</sup> and (b) from 1725 to 1625 cm<sup>-1</sup>.



**Fig. S11** FT-IR spectra ranged (a) from 3300 to 3150 cm<sup>-1</sup> and (b) from 1725 to 1625 cm<sup>-1</sup> of **DPCHP-DODE** at 40 °C cooled from the melt in KBr sheet.

## References

S1 AIST: Integrated Spectral Database System of Organic Compounds. (Data were obtained from the National Institute of Advanced Industrial Science and Technology (Japan)).