

Supporting Information

Special interplay of hydrogen bonds and dynamic covalent bonds in sustainable polyurethane vitrimers with excellent recyclability and reprocessability

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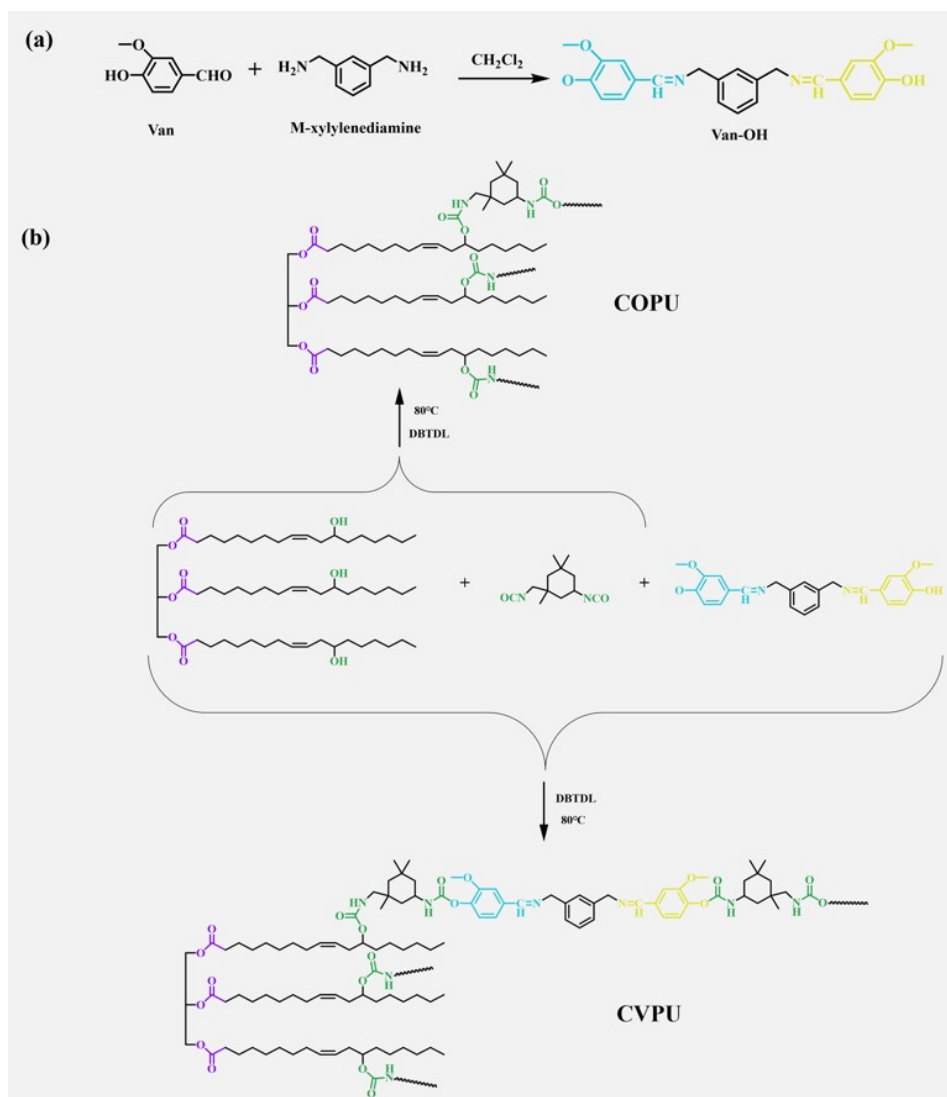


Fig. S1 (a) The synthesis route of the Van-OH. (b) The synthesis route of the COPU and the PU vitrimers (the CVPU).

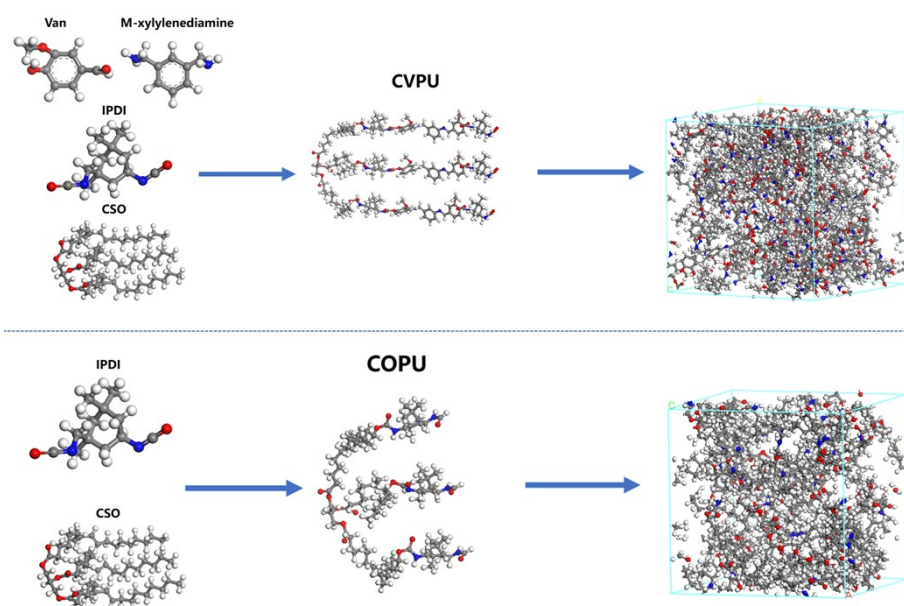


Fig. S2 Construction of the simulated CVPU system (the upper part) and the simulated COPU system (the lower part).

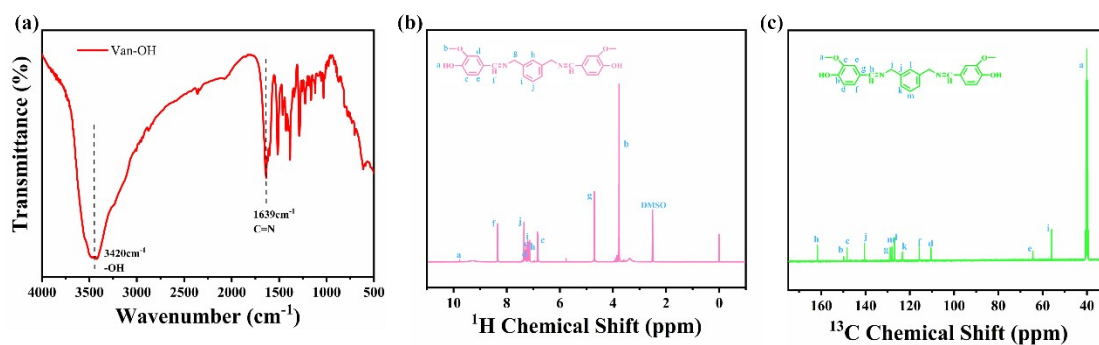


Fig. S3 (a) FTIR spectrum, (b) ^1H NMR spectrum and (c) ^{13}C NMR spectrum of the Van-OH.

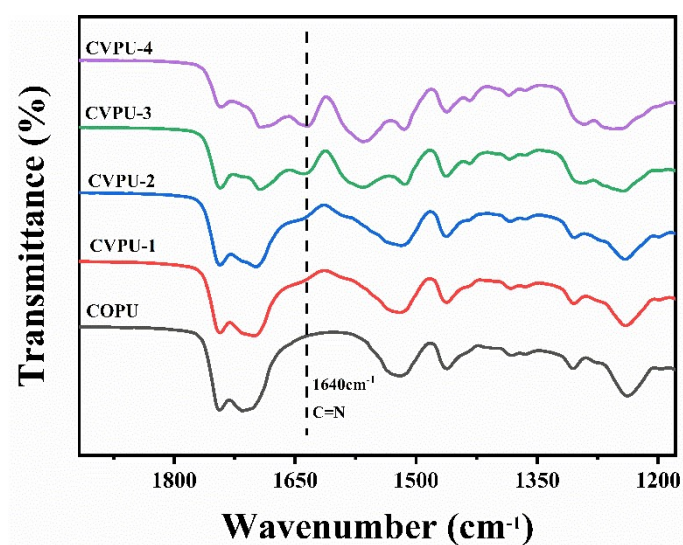


Fig. S4 Localized magnification of FTIR spectra of the COPU and CVPU samples.

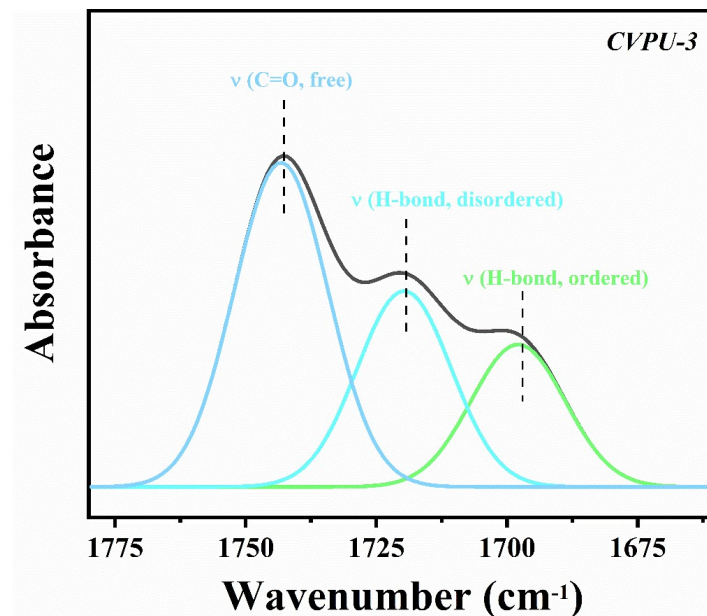


Fig. S5 The absorption peak corresponding to the C=O stretching vibrations which can be divided into three sub-peaks.

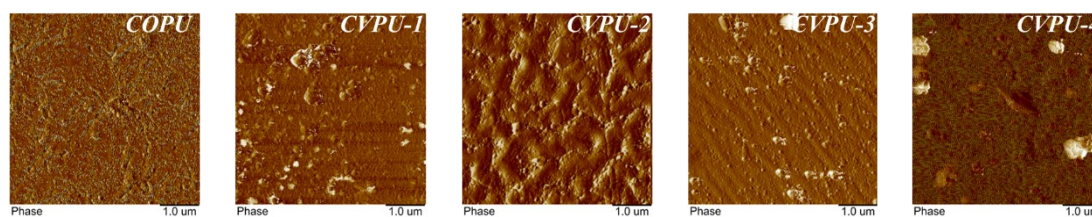


Fig. S6 AFM images of the COPU, CVPU-1, CVPU-2, CVPU-3, and CVPU-4 samples.

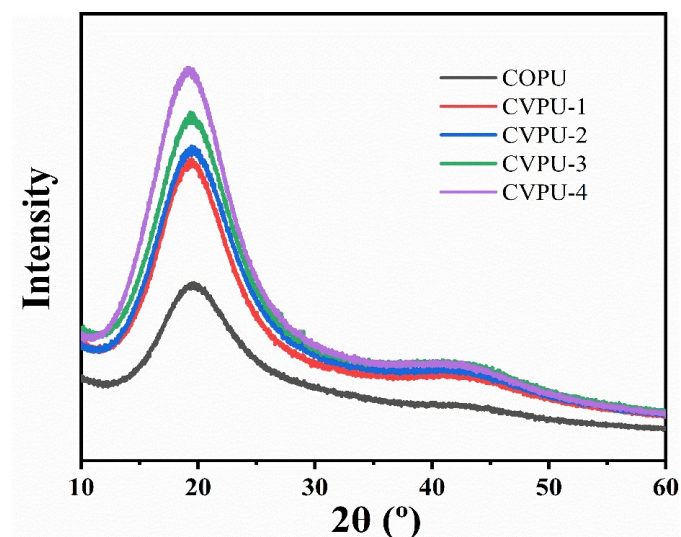


Fig. S7 XRD patterns of the COPU and CVPU-1/2/3/4 samples.

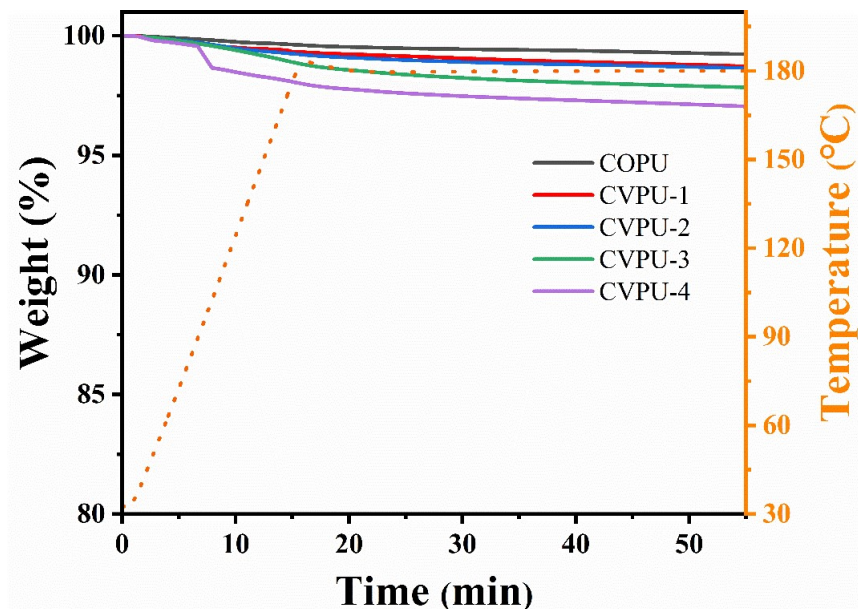


Fig. S8 Isothermal TGA curves of five samples at 180°C for 40 min.

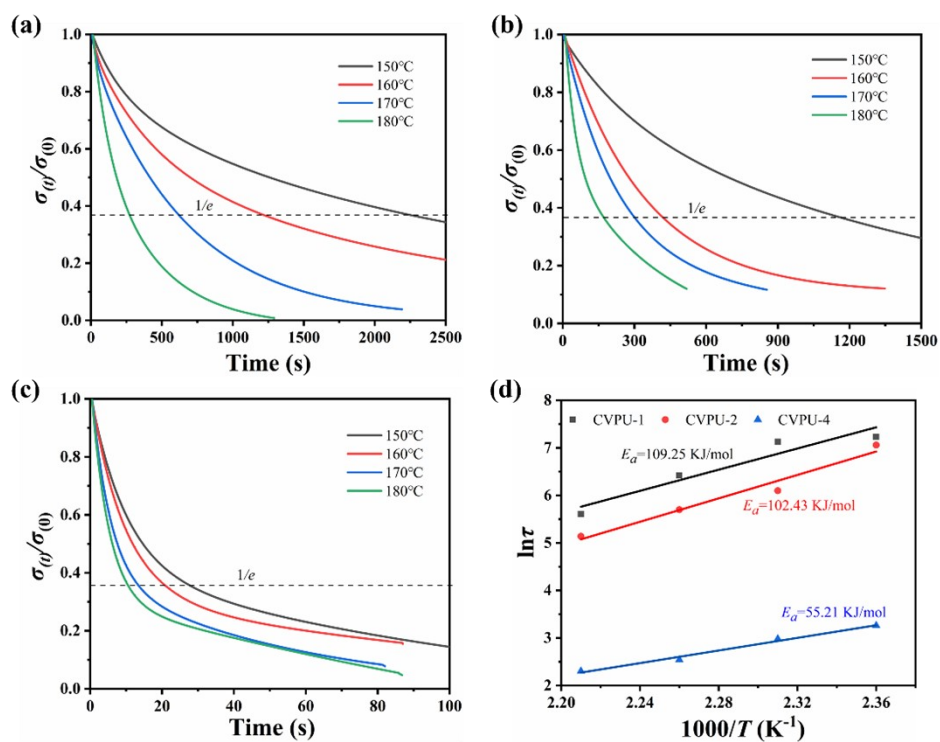


Fig. S9 Changes of the $\sigma(t)/\sigma(0)$ of (a) the CVPU-1 sample, (b) the CVPU-2 sample, and (c) the CVPU-4 sample as a function of time during stress relaxation at different temperatures. (d) $\ln \tau$ as a

function of $1000/T$ for the CVPU-1 sample, the CVPU-2 sample and the CVPU-4 sample (the solid lines were obtained by fitting the data based on the Arrhenius equation).

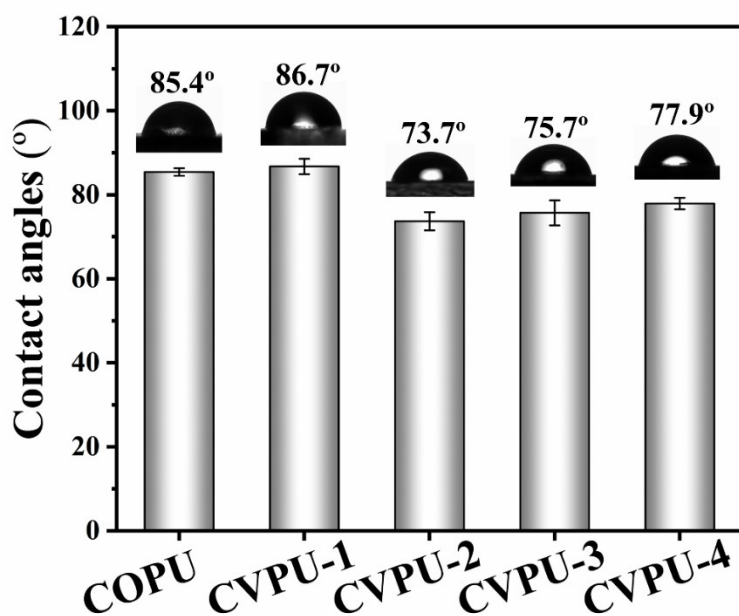


Fig. S10 The water contact angle of the COPU sample and the CVPU-1/2/3/4 samples based on the water contact angle test.

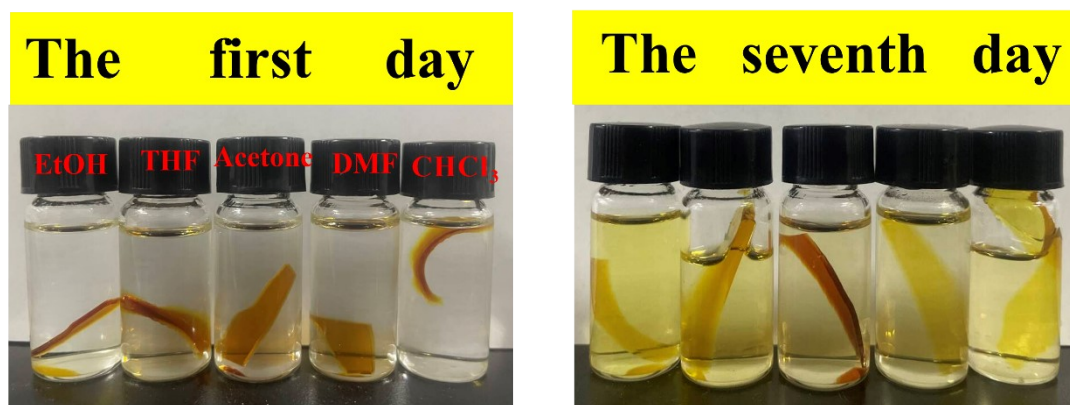


Fig. S11 Photos of the CVPU-3 samples soaked in the solvents for different durations.

Table S1 Formulation of the different PU samples

Sample	COPU	CVPU-1	CVPU-2	CVPU-3	CVPU-4
Van-OH (mol)	0	0.04	0.0085	0.0125	0.0185
CSO (mol)	0.01	0.01	0.01	0.01	0.01

IPDI (mol)	0.01575	0.0209	0.02585	0.03025	0.0737
DBTDL (g)	0.13	0.156	0.185	0.2	0.25

Table S2 Crosslinking density for the COPU and CVPU-3/4 samples

Sample	COPU	CVPU-3	CVPU-4
Crosslinking density*10⁻⁴ (mol/ml)	92.6	86.4	84.4

Table S3 Thermal stability data of the COPU sample and the five CVPU samples

	T_5 (°C)	T_{max1} (°C)	T_{max2} (°C)
COPU	309.4	335.7	436.6
CVPU-1	297.2	335.3	428.9
CVPU-2	240.7	338.5	437.7
CVPU-3	205.4	338.3	423.7
CVPU-4	191.8	337.5	415.1

T_5 represents the temperature at which the quality of the sample has decreased by 5%.

T_{max1} and T_{max2} respectively represent the two temperatures at which the values of weight loss rate are greatest in the DTG curves.

Table S4 The R^2 values corresponding to the fitting of the Arrhenius plots

Sample	CVPU-1	CVPU-2	CVPU-3	CVPU-4
R^2	0.91775	0.96642	0.89736	0.98767

Table S5 Tensile strength, elongation at break and Young's modulus of the five PU samples

	Tensile strength	Elongation at	Young's modulus	Toughness
	(MPa)	break (%)	(MPa)	(MJ/m³)
COPU	1.47	384.74	4.39	2.93
CVPU-1	15.26	428.53	60.91	25.69
CVPU-2	24.73	526.89	191.68	64.11
CVPU-3	28.43	400.62	597.99	68.75
CVPU-4	36.69	238.52	1751.83	66.64

Table S6 Hysteresis loop areas of the cyclic stress-strain curves for the COPU and the different CVPU samples

Sample	COPU	CVPU-1	CVPU-2	CVPU-3	CVPU-4
Area (MJ/m³)	0.2	3.8	7.1	15.4	33.7