## **Electronic supplementary information (ESI) for**

## Meldrum's acid mediated ketene chemistry in formation of ester bonds for synthesis of vitrimers with high glass transition temperatures

Du-Yuan Hung and Ying-Ling Liu\*

Department of Chemical Engineering, National Tsing Hua University, No. 101, Sec. 2, Kuang-Fu Road, Hsinchu 300044, Taiwan.









**Figure S1**. Spectral characterization of MADV with (a) FTIR, (b) <sup>1</sup>H NMR, (c) <sup>13</sup>C NMR, and (d) molecular mass instruments. It is noted that the acetone signals could be from the residual solvent employed in the synthesis and purification processes.



*Figure S2.* FTIR spectra of MADV recorded at different temperatures for tracing the occurrence of MA thermolysis reaction.



*Figure S3.* GPC chromatograms of PMADV-6 and PMADV-12 for determination their molecular weights.



*Figure S4*. Storage modulus and loss modulus of PMADV and CR-MADV recorded at 150 °C and various frequencies.



Figure S5. Stress relaxation curves of CR-MADV recorded at various temperatures.



**Figure S6**. Stress relaxation curves with curve fitting (upper) and Arrhenius plots (lower) of CR-MADV and R-CR-MADV-3.







*Figure S7*. (a) SEM micrographs and (b) stress-strain curves tracing the mendable tests of CR-MADV at 230 °C under an applied pressure of about of about 34,000 N m<sup>-</sup><sup>2</sup>. The SEM pictures in relatively high magnification exhibiting the mended cutprint of the sample.



**Figure S8**. SEM microgrphas of the recycled sample of CR-MADV obtained from thermal press at 200 °C and 5 MPa for 1 h.



*Figure S9*. FTIR measurements on CR-MADV isothermally at 230 °C, indicating no obvious changes in the chemical structure being observed in the duration of reprocessing.



*Figure S10*. TGA thermograms of CR-MADV and the thermally recycled samples (R-CR-MADV), indicating no obvious changes in thermal stability of CR-MADV after being thermally recycled.



*Figure S11*. Stress-strain curves of CR-MADV and the thermally recycled samples (R-CR-MADV.



*Figure S12. (a)* Stress-strain curves and (b) TGA thermograms of pristine, solvent-(NMP), acid-, and base-treated CR-MADV samples.

Sample	Young's modulus (GPa)	Tensile strength (MPa)	Elongation at break (%)	Recycling efficiency (%)
CR-MADV	1.58	28.6	2.4	
R-CR-MADV-1	1.48	24.0	2.0	84
R-CR-MADV-2	1.78	22.5	1.5	94
R-CR-MADV-3	1.22	15.8	1.8	70

**Table S1**. Mechanical properties of CR-MADV and the thermallyrecycled samples.

**Table S2**. Gel fractions (%) of CR-MADV and the thermallyrecycled samples.

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Sample	Gel fraction (wt%)			
	NMP	1 N HCl <sub>(aq)</sub>	1 N NaOH <sub>(aq)</sub>	
CR-MADV	99.1	97	96.8	
R-CR-MADV-1	99.4	96.8	98.4	
R-CR-MADV-2	98.9	96.2	98.4	
R-CR-MADV-3	97.8	95.2	98.5	