Supporting Information For

Fusion of biobased vinylogous urethane vitrimers with distinct mechanical

properties

Fengbiao Chena, Fei Gao*a, Jiang Zhonga, Liang Shen*a, Yangju Lin*b

^aJiangxi Engineering Laboratory of Waterborne Coating, School of Chemistry and Chemical Engineering, Jiangxi Science &Technology Normal University, Nanchang 330013, Jiangxi, P. R.

China

^bDepartment of Chemistry, Duke University, Durham, North Carolina 27708, United States

E-mail address: feigao2016@jxstnu.com.cn (F. Gao);liangshen@jxstnu.com.cn(L. Shen).

yangju.lin@duke.edu (Y, Lin)

Table of Contents

1. Calculating the yield of acetoacetate-modified cardanol (AMCA) by ¹ H NMR	2
1.1 Yield of acrylate cardanol (ACA)	2
1.2 Yield of modified cardanol (MCA)	2
1.3 Yield of acetoacetate-modified cardanol (AMCA)	3
2. Mechanical and thermal properties of polymer networks	3
3. Reprocessing of AMCA-M and AMCA-P polymer networks	4
3.1 Characterization of remolded of AMCA-M polymer networks	4
3.2 Characterization the remold reprocessing of AMCA-P networks	5
3.3 The DSC overlay of 2nd remolded AMCA-MPS and AMCA-MP	6
4. Dissolving behavior of remolded AMCA-MPS polymer network in xylenes, cyclohexylamine a	and
benzylamine	6

1. Calculating the yield of acetoacetate-modified cardanol (AMCA) by ¹H NMR



1.1 Yield of acrylate cardanol (ACA)

Figure S1. ¹H NMR spectra of (a) cardanol (CA) and (b) acrylate cardanol (ACA)

Figure S1a shows that the peak areas of a', b', c', and d' are 4, and the peak area of g' is 0.42, the peak area of f' is 3.28, the peak area of h' is 0.88 and the peak area of q' is 1.77; thus, the ¹H NMR peak areas correspond to the compound structure. Therefore, we can calculate the conversion yield on the basis of the peak area ratios. the yield of acrylate cardanol (ACA) can be calculated as: W(%) = A(r+s+q)/3=(0.99+0.98+0.99)/3=99%.

1.2 Yield of modified cardanol (MCA)



Figure S2. ¹H NMR spectra of (a) acrylate cardanol (ACA) and (b) modified cardanol (MCA) The yield of modified cardanol is calculated using the same method as described in section 1.1:

(MCA) W(%) = [A(r+s+q+f+e+g) - A(m')] / A(r+s+q+f+e+g) = [7.56 - 0.42] / 7.88 = 95 %.



1.3 Yield of acetoacetate-modified cardanol (AMCA)

Figure S3. ¹H NMR spectra of acetoacetate-modified cardanol (AMCA).

Using the same method as mentioned in section 1.1, the yield of acetoacetate-modified cardanol (AMCA) is: W% = A(i) / [A(i)+A(i')] = 5.65 / [5.65+0.41] = 93 %. And if we set the peak area of h is 1.77, the peak of g is 5.41, that means there is about 2.7 acetoacetate groups.

2. Mechanical and thermal properties of polymer networks

	Young's	Stress at	Elongation at Break (%)	DSC T _g (°C)	TGA in nitrogen (°C)		
Sample	modulus (MPa)	Break (MPa)			T ₁₀	T ₅₀	T _{max}
AMCA-M	14.8 ± 0.3	3.30 ± 0.07	112 ± 3.0	18	251	351	451

Table S1. Mechanical and thermal properties of polymer networks

AMCA-P	$74.3~{\pm}4.0$	7.51 ± 0.13	31 ± 0.86	35	263	345	456
AMCA-MP	33.3 ± 1.5	6.48 ± 0.02	152 ± 0.67	31	266	349	454

T10, T50, and T_{max} represent the temperatures for a mass loss of 10 *wt* %, 50 *wt* %, and the maximum mass loss, respectively.

3. Reprocessing of AMCA-M and AMCA-P polymer networks

3.1 Characterization of remolded of AMCA-M polymer networks



Figure S4. Characterization of AMCA-M polymer networks before and after reprocessing using (a) FTIR spectra analysis, (b) tensile tests, (c) TGA analysis, and (d) DSC measurement.



3.2 Characterization the remold reprocessing of AMCA-P networks

Figure S5. Characterization of AMCA-P polymer networks before and after reprocessing using (a) FTIR spectra analysis, (b) tensile tests, (c) TGA analysis, and (d) DSC measurement.

3.3 The DSC overlay of 2nd remolded AMCA-MPS and AMCA-MP



Figure S6. the DSC overlay of 2nd remolded AMCA-MPS and AMCA-MP

4. Dissolving behavior of remolded AMCA-MPS polymer network in xylenes, cyclohexylamine and benzylamine.



Figure S7. AMCA-MPS polymer network was subjected to two rounds of reprocessing and dissolved in xylenes, cyclohexylamine and benzylamine at 130 °C for 40 min.



Figure S8. ¹H NMR Spectrum of CA (400 MHz, CDCl₃, 7.26 ppm)



Figure S9. ¹H NMR Spectrum of ACA (400 MHz, CDCl₃, 7.26ppm)



Figure S11. ¹H NMR Spectrum of MCA (400 MHz, CDCl₃, 7.26 ppm)



Figure S12. ¹³C NMR Spectrum of MAC (400 MHz, CDCl₃, 77 ppm)



Figure S13. ¹H NMR Spectrum of AMCA (400 MHz, CDCl₃, 7.26 ppm)



Figure S14. ¹³C NMR Spectrum of AMCA (400 MHz, CDCl₃, 77 ppm)