ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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Vinylogous Urethane Vitrimers

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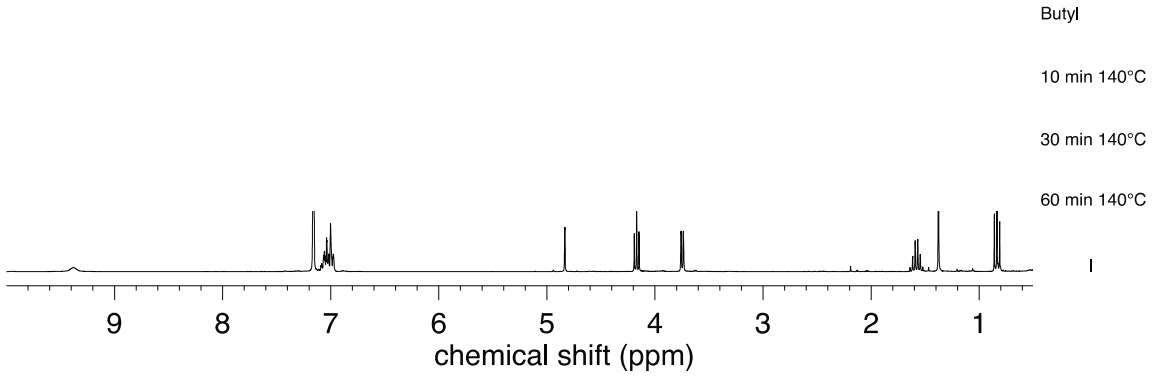

Figure S1: NMR spectra of a pure *N*-butyl vinylogous urethane (upper), *N*-benzyl vinylogous urethane (lower) and of the resulting mixture after 10, 30 and 60 minutes at 140°C.

Calculation activation energy

The decrease of the reactant as a function of time is described by [28]:

$$[Reactant] = 1 - (x_\infty - \exp\left(\frac{-kt}{x_\infty}\right))$$

$$\text{With } x_\infty = \text{equilibrium concentration of the products} = \frac{5}{6}$$

$$Fitted\ for\ k = initial\ rate$$

The activation energy was calculated by plotting ln k versus 1000/T.

$$\ln k = -\frac{E_a}{RT} + cte$$

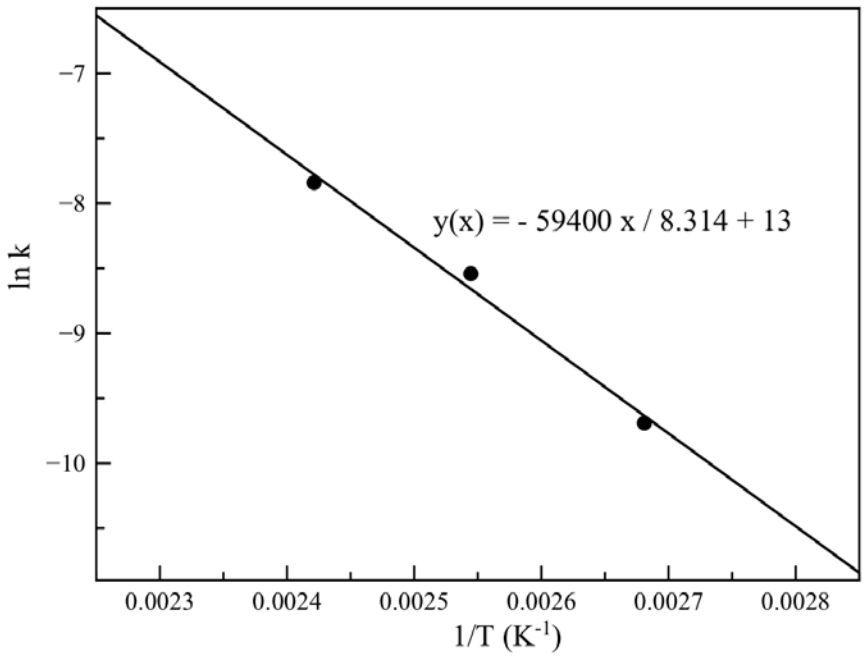

Figure S2: The activation energy was calculated by plotting lnk vs 1/T.

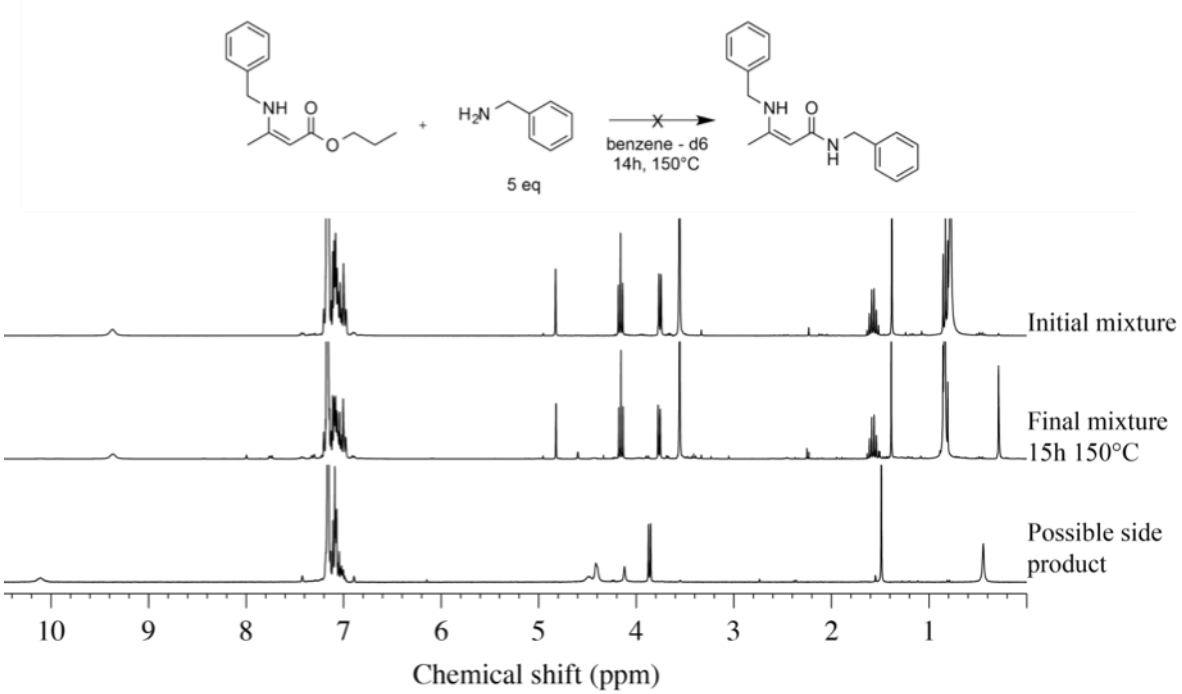

Figure S3: Control experiment to discard the occurrence of side reactions.

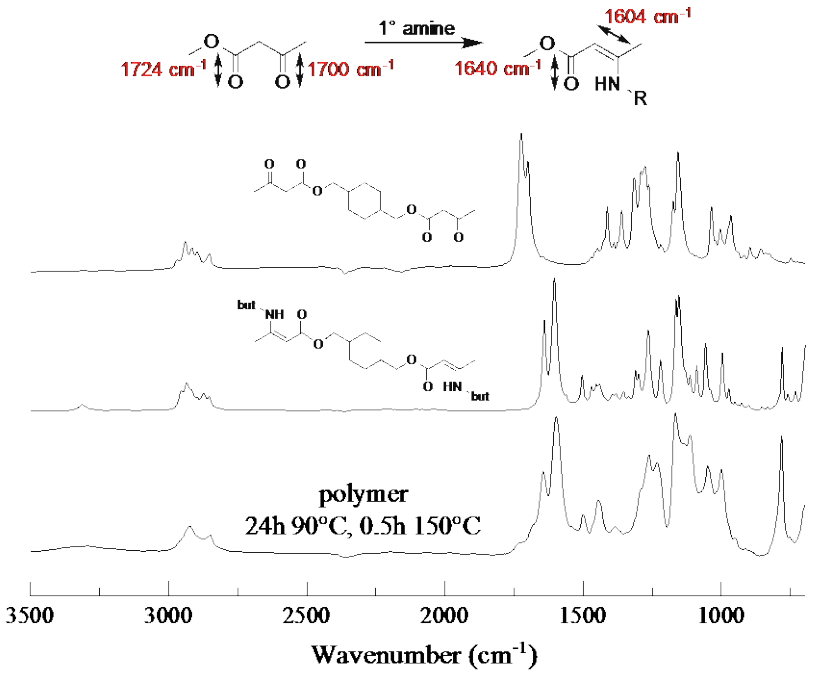

Figure S4: Conversion of the acetoacetate function to vinylogous urethanes followed by FTIR. After curing, close to full conversion was observed.

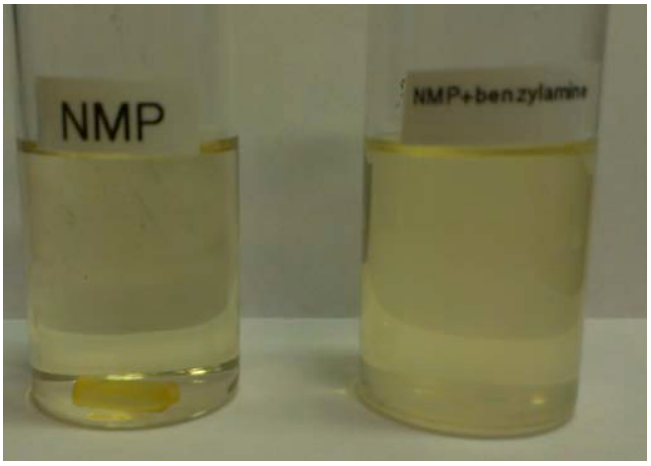

Figure S5: Solubility test on the poly(vinylogous urethane) network: the network is insoluble after 24h at 100°C in NMP (left). Upon addition of benzyl amine, the network dissolves as a result of exchange reactions (right).

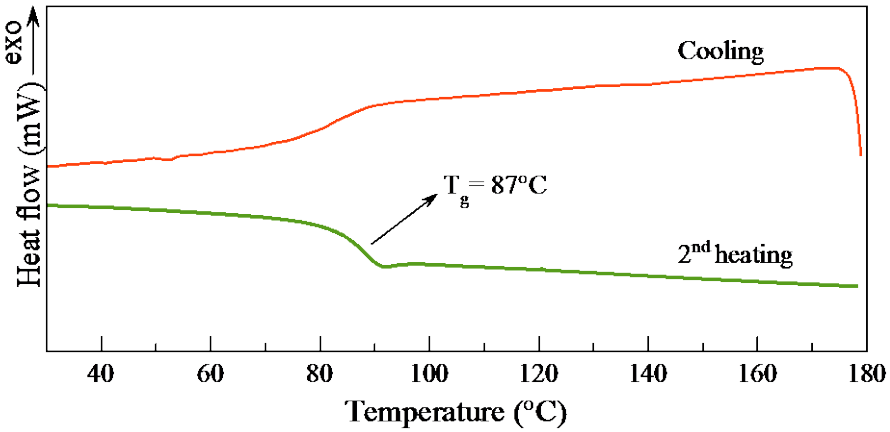

Figure S6: DSC thermogram of the poly(vinylogous urethane) network.

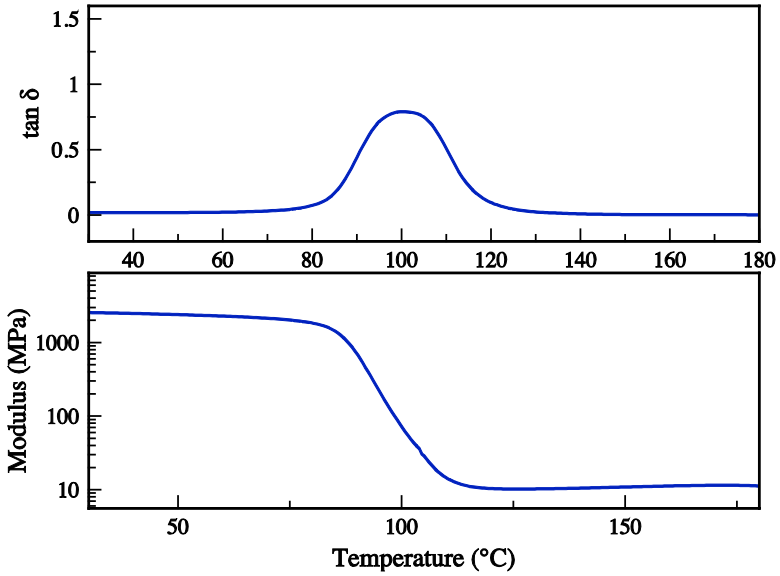

Figure S7: DMTA of the poly(vinylogous urethane) network. A modulus of 2.4 GPa and a rubbery plateau of 10 MPa were measured.

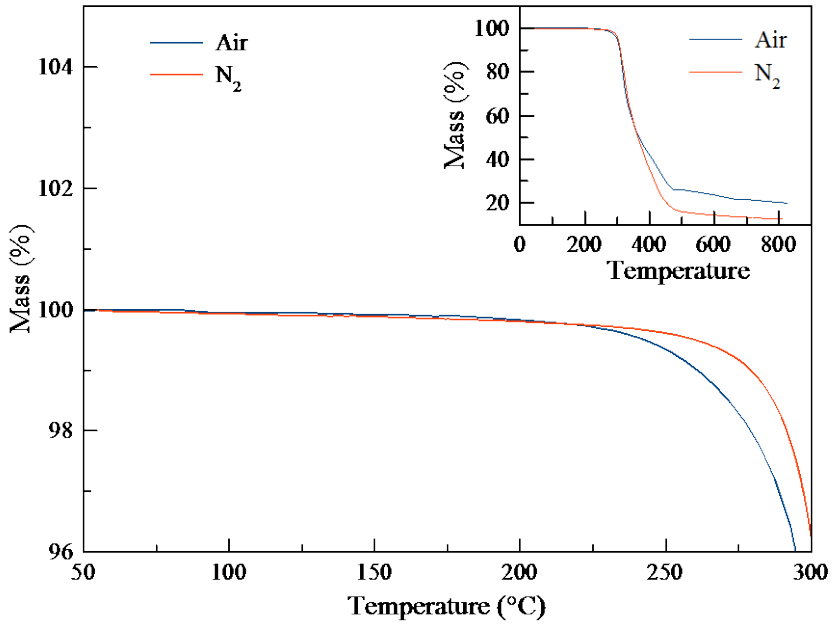

Figure S8: TGA of the poly(vinylogous urethane) network under air and nitrogen atmosphere.

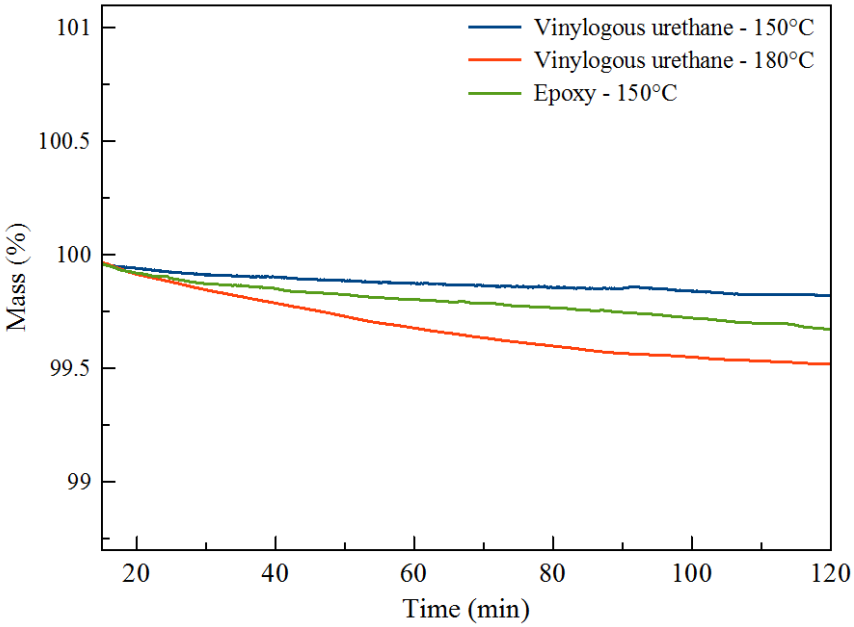

Figure S9: Isothermal TGA of the poly(vinylogous urethane) network at 150°C and 180°C. The isothermal TGA of a commercial epoxy (EPON 828 cured with DETA) at 150°C is shown as a reference.

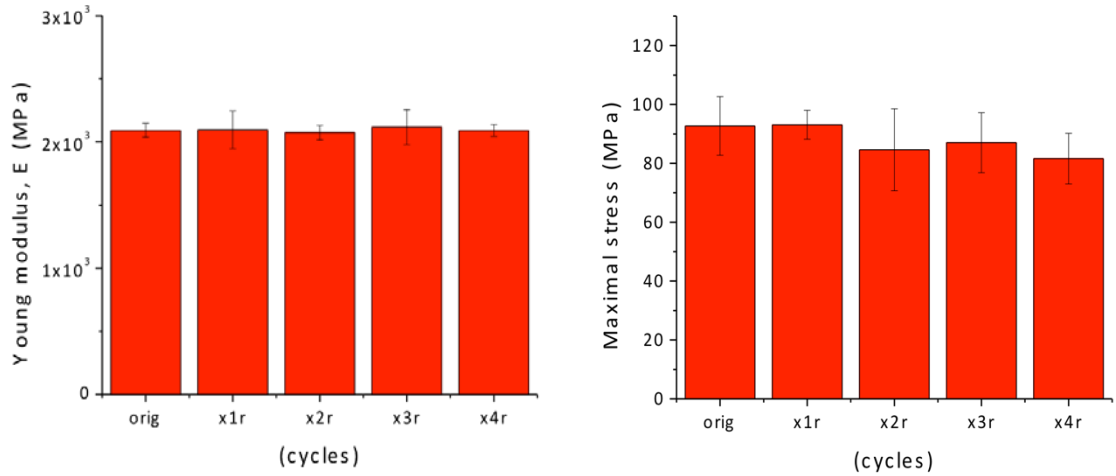

Figure S10: Young modulus and maximal stress of the poly(vinylogous urethane) network before and after up to four consecutive recycling cycles.

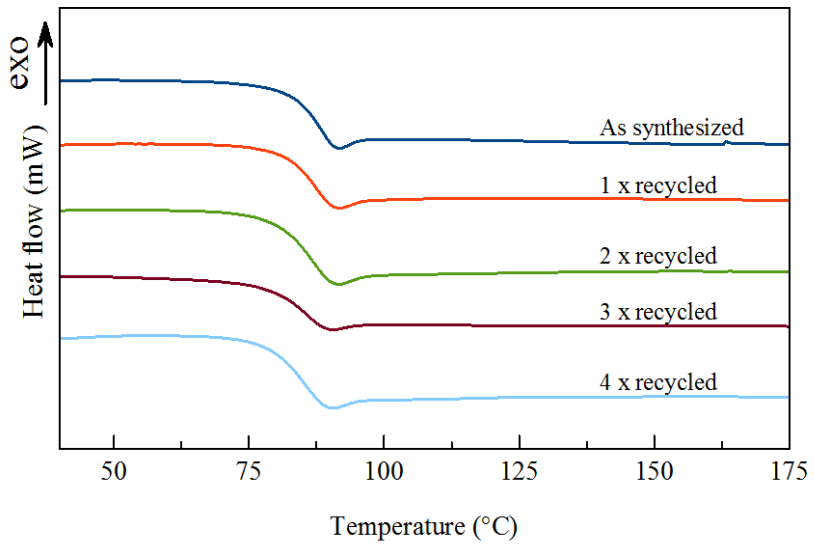

Figure S11: DSC thermograms of the poly(vinylogous urethane) network before and after up to four consecutive recycling cycles.

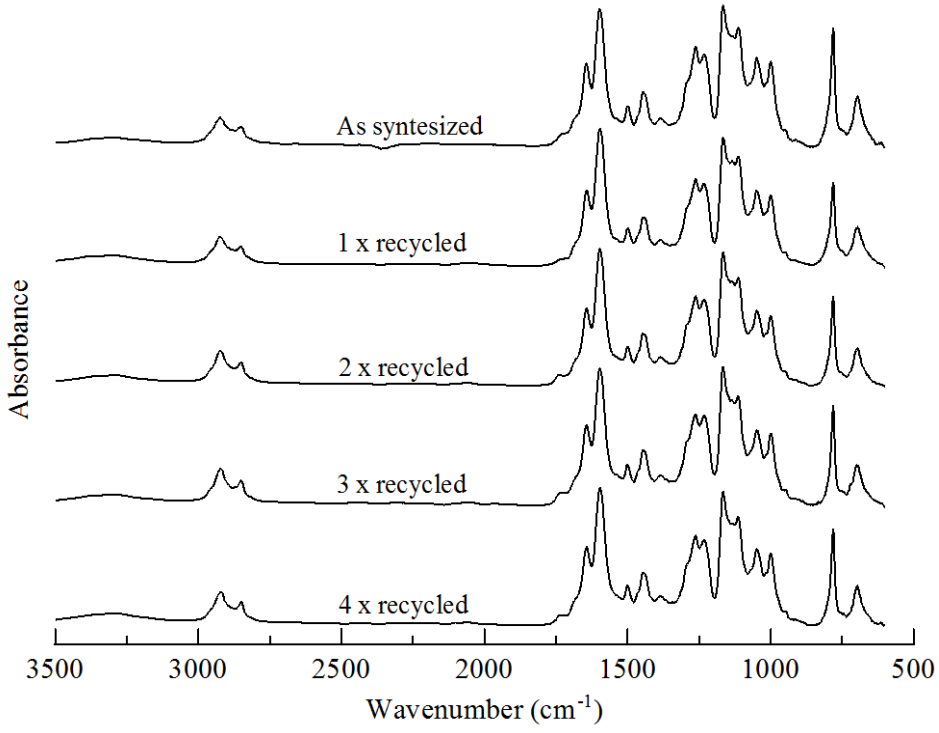

Figure S12: FT-IR spectra of the poly(vinylogous urethane) network before and after up to four consecutive recycling cycles.

Table S1: Soluble fraction of the original poly(vinylogous urethane) network and the recycled samples.

Sample	Soluble fraction (%)
As synthesized	9 - 13
1 x recycled	6 - 8
2x recycled	9 - 11
3x recycled	12 - 15
4x recycled	8 - 12

[a)] The lowest and highest value of three measurements are shown.

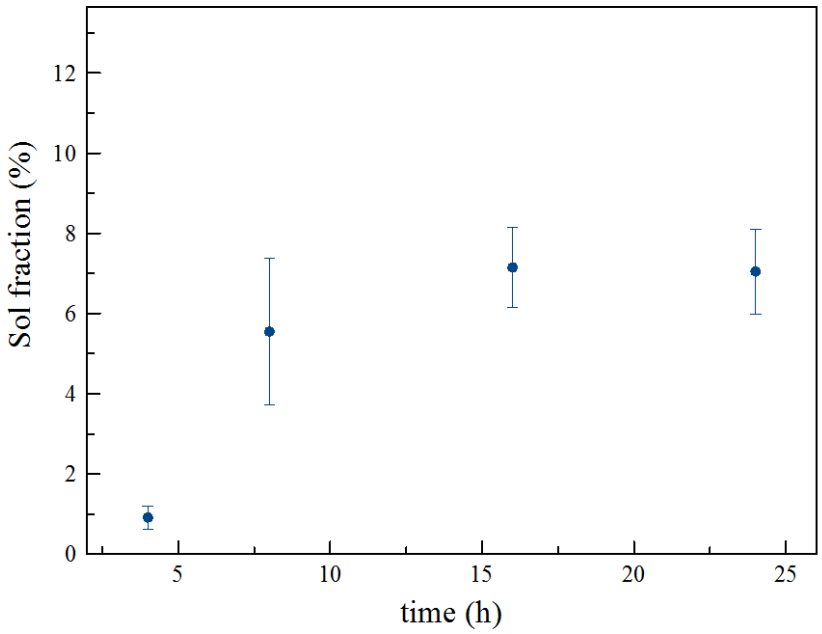

Figure S13 Sol fraction of a 1x recycled sample at 100°C in NMP as a function of time.