

ELECTRONIC SUPPLEMENTARY INFORMATION

**Poly(*N*-vinylimidazole)-/*l*-poly(propylene glycol) amphiphilic conetworks and gels:
molecularly forced blends of incompatible polymers with single glass transition
temperatures of unusual dependence on composition**

Csaba Fodor,^{a,*†} Tímea Stumphauser,^a Ralf Thomann,^b Yi Thomann^b and Béla Iván^{a*}

^aPolymer Chemistry Research Group, Institute of Materials and Environmental Chemistry,
Research Centre for Natural Sciences, Hungarian Academy of Sciences, H-1117 Budapest,
Magyar tudósok krt. 2., Hungary

^bFreiburg Materials Research Center, Albert-Ludwigs University Freiburg,
Stefan-Meier-Str. 21, D-79104 Freiburg, Germany

e-mail addresses:	Cs. Fodor*:	c.fodor@rug.nl
	T. Stumphauser:	stumphauser.timea@ttk.mta.hu
	R. Thomann:	ralf.thoman@fmf.uni-freiburg.de
	Y. Thomann:	yi.thomann@fmf.uni-freiburg.de
	B. Iván*:	ivan.bela@ttk.mta.hu

Present address: [†] Department of Polymer Chemistry, Zernike Institute for Advanced
Materials, University of Groningen
Nijenborgh 4, 9747 AG Groningen, The Netherlands

* Authors to whom correspondence should be addressed.

The number average molecular weight (M_n) of the poly(propylene glycol) dimethacrylate (PPGDMA) macromolecular cross-linker was determined by ^1H NMR and by gel permeation chromatography (GPC) measurements.

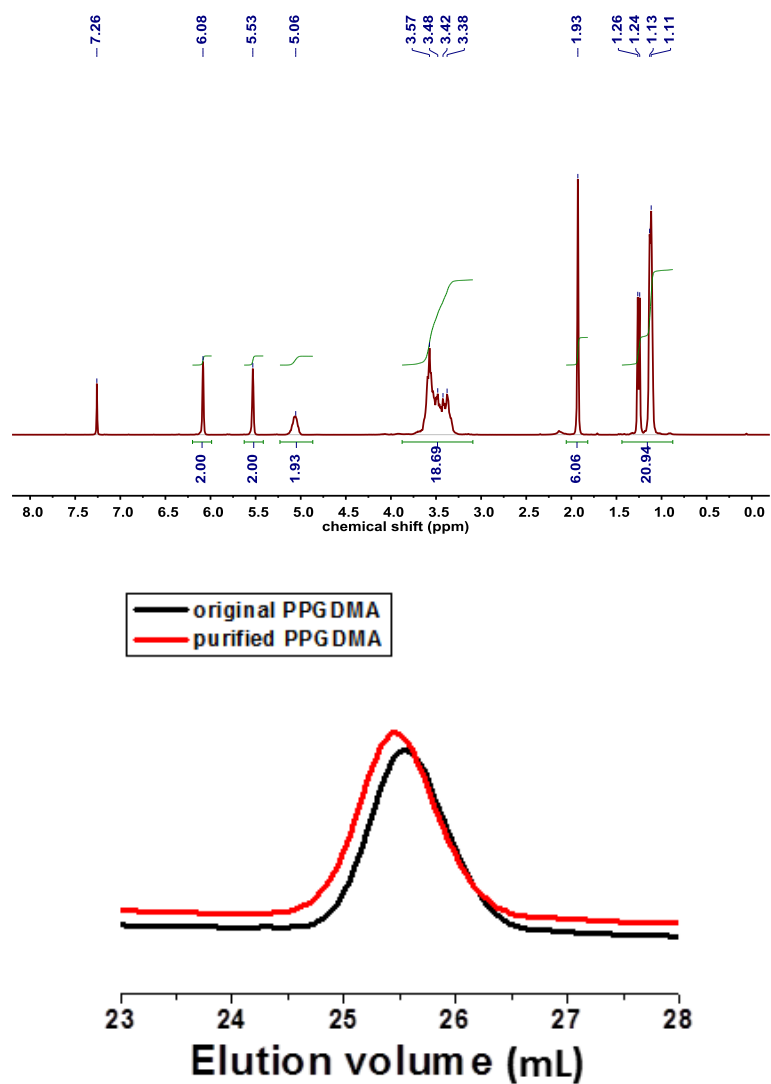


Figure S1. ^1H NMR spectrum (upper) of the poly(propylene glycol) dimethacrylate (PPGDMA) macromolecular cross-linker, recorded in CDCl_3 at room temperature and GPC traces of the original and the inhibitor remover treated sample (lower).

For comparison with the conetworks, polymer blends composed of poly(*N*-vinylimidazole) (PVIIm) homopolymer and PPGDMA macromonomer with different weight ratios were prepared by solvent casting from common solvent of ethanol (Table S1 and Figure S2) to investigate and compare the thermal behavior and stability of the not cross-linked immiscible polymer systems (Figure S3 and Table S2).

Table S1. Weight ratios of poly(*N*-vinylimidazole) (PVIIm) homopolymer and poly(propylene glycol) dimethacrylate (PPGDMA) macromonomer ($M_n = 560 \text{ g}\cdot\text{mol}^{-1}$) in their mixtures with different PVIIm/PPGDMA ratios (20, 40, 60 and 80 wt% of PPGDMA).

Sample ID	PVIIm/PPG in feed (wt%)	m(PPGDMA) (mg)	m(PVIIm) (mg)	m(blend) (mg)	V(EtOH) (mL)
B-20	80/20	40.0	159.3	199.3	1.96
B-40	60/40	79.9	120.4	200.3	1.92
B-60	41/59	120.0	84.5	204.5	1.88
B-80	21/79	160.0	42.5	202.5	1.84



Figure S2. Photographs of poly(*N*-vinylimidazole) (PVIIm) and poly(propylene glycol) dimethacrylate (PPGDMA) homopolymer blends (PVIIm-*blend*-PPG) with different compositions: 20 wt% (a), 40 wt% (b), 60 wt% (c) and 80 wt% of PPG (d).

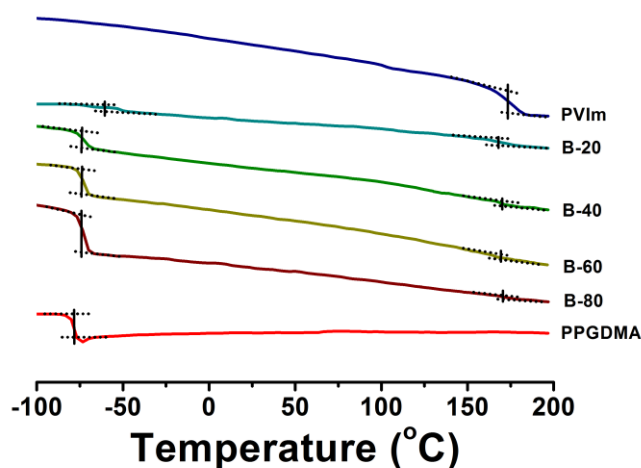


Figure S3. Differential scanning calorimetry (DSC) thermograms (second scan) of poly(*N*-vinylimidazole) and poly(propylene glycol) homopolymer blends (PVIm-*blend*-PPG), PVIm homopolymer and PPGDMA macromolecular cross-linker. Glass transition temperatures (T_g) are represented by ticks.

Table S2. Glass transition temperature (T_g) values of poly(*N*-vinylimidazole) (PVIm) and poly(propylene glycol) dimethacrylate (PPGDMA) homopolymer blends (PVIm-*blend*-PPG), PVIm homopolymer and PPGDMA cross-linker ($M_n = 560 \text{ g}\cdot\text{mol}^{-1}$), respectively.

Sample ID	PVIm/PPG in feed (wt%)	T_{g1} (°C)	T_{g2} (°C)
B-20	80/20	-60	167
B-40	60/40	-73	169
B-60	41/59	-74	169
B-80	21/79	-74	170
PVIm	100/0	-	171
PPGDMA	0/100	-76	-

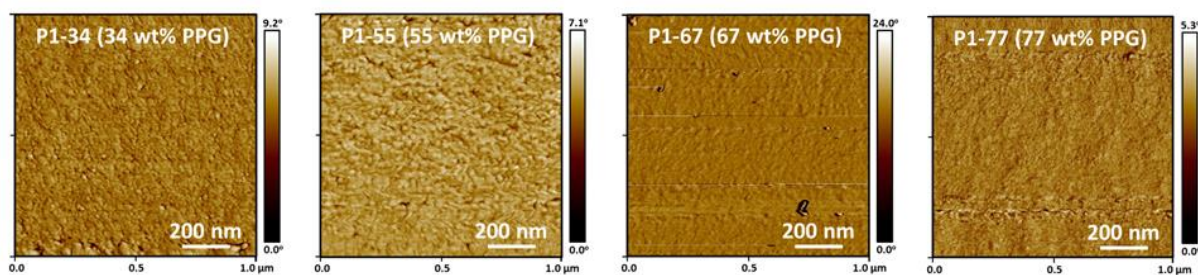


Figure S4. Atomic force microscopy (AFM) phase mode images of the cross sections (bulk morphology) of poly(*N*-vinylimidazole)-*l*-poly(propylene glycol) (PVI*m-l*-PPG) conetwork varying PPG content (35 wt%, 55 wt%, 67 wt% and 77 wt% PPG) (picture dimensions 1 μm x 1 μm). By minimizing the phase range of the vertical scale bar, the color contrast was maximized such that the sectioning structure, based on topography difference instead of on material difference, was observed. This is proven by the horizontal sectioning direction, which is visible in all images.

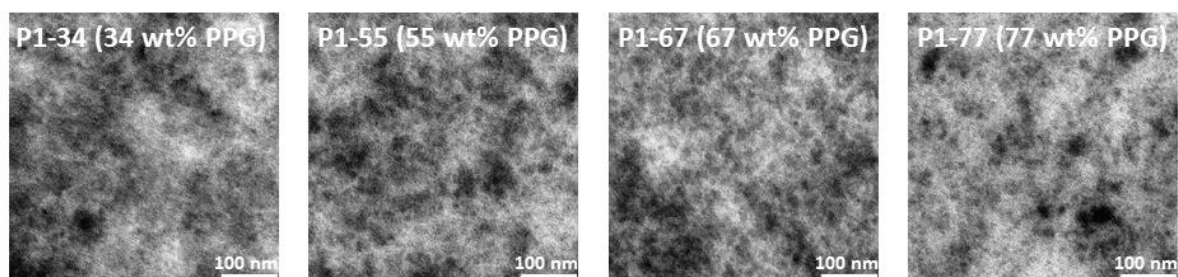


Figure S5. Transmission electron microscopic (TEM) pictures of poly(*N*-vinylimidazole)-*l*-poly(propylene glycol) (PVI*m-l*-PPG) conetwork samples with varying PPG content (35 wt%, 55 wt%, 67 wt% and 77 wt% PPG) stained with OsO_4 (picture dimensions 500 nm x 500 nm).

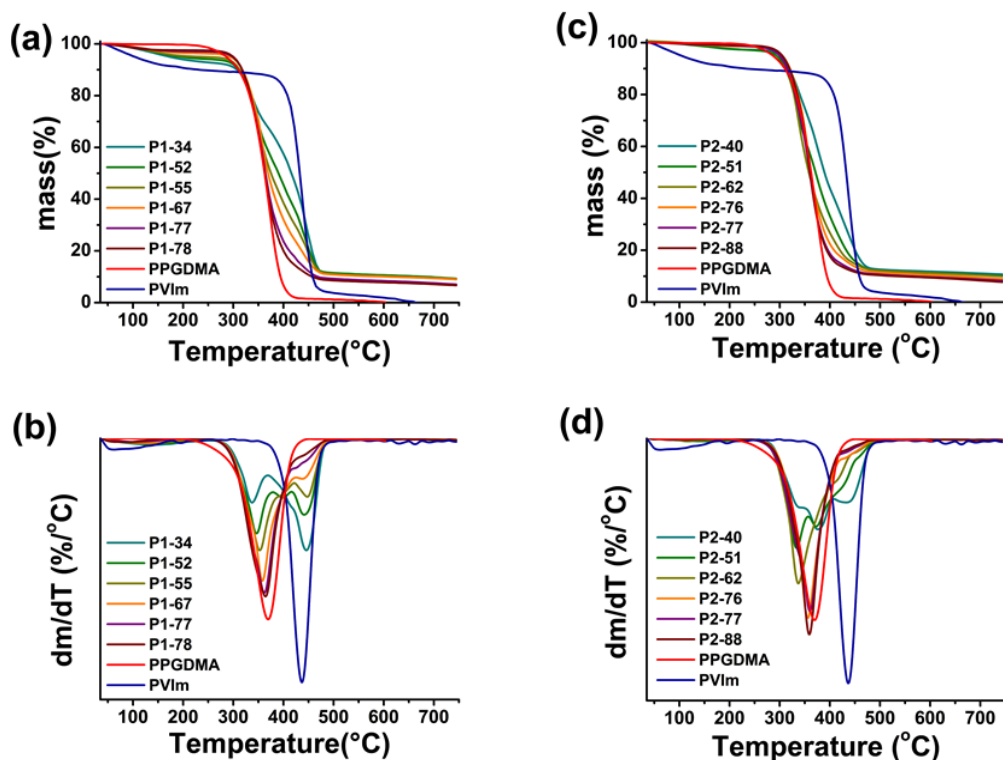


Figure S6. Thermogravimetric analysis (TGA) (a and c) and differential TGA (DTG) (b and d) curves of poly(*N*-vinylimidazole)-*l*-poly(propylene glycol) (PVIIm-*l*-PPG) conetworks, PVIIm homopolymer and PPGDMA macromonomer obtained with $10\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ heating rate under nitrogen.

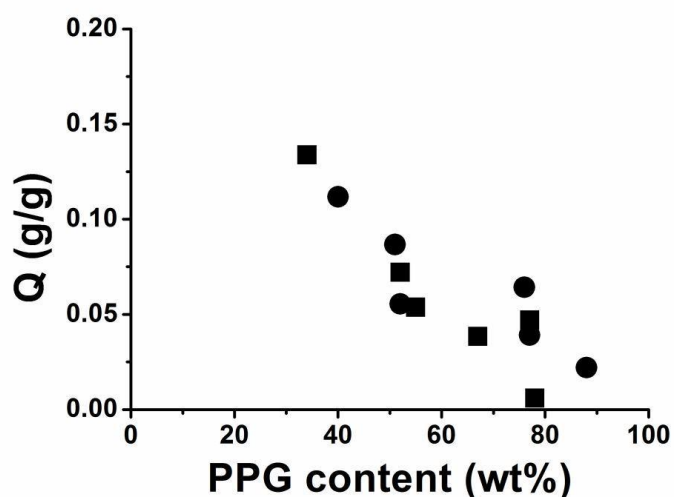


Figure S7. Equilibrium water adsorption (hygroscopic swelling ratios) (Q) of the poly(*N*-vinylimidazole)-*l*-poly(propylene glycol) (PVIIm-*l*-PPG) conetworks ((■) P1 and (●) P2 PVIIm-*l*-PPG conetwork series) as a function of the PPG content at ambient conditions.