

Supporting Information:

Methods

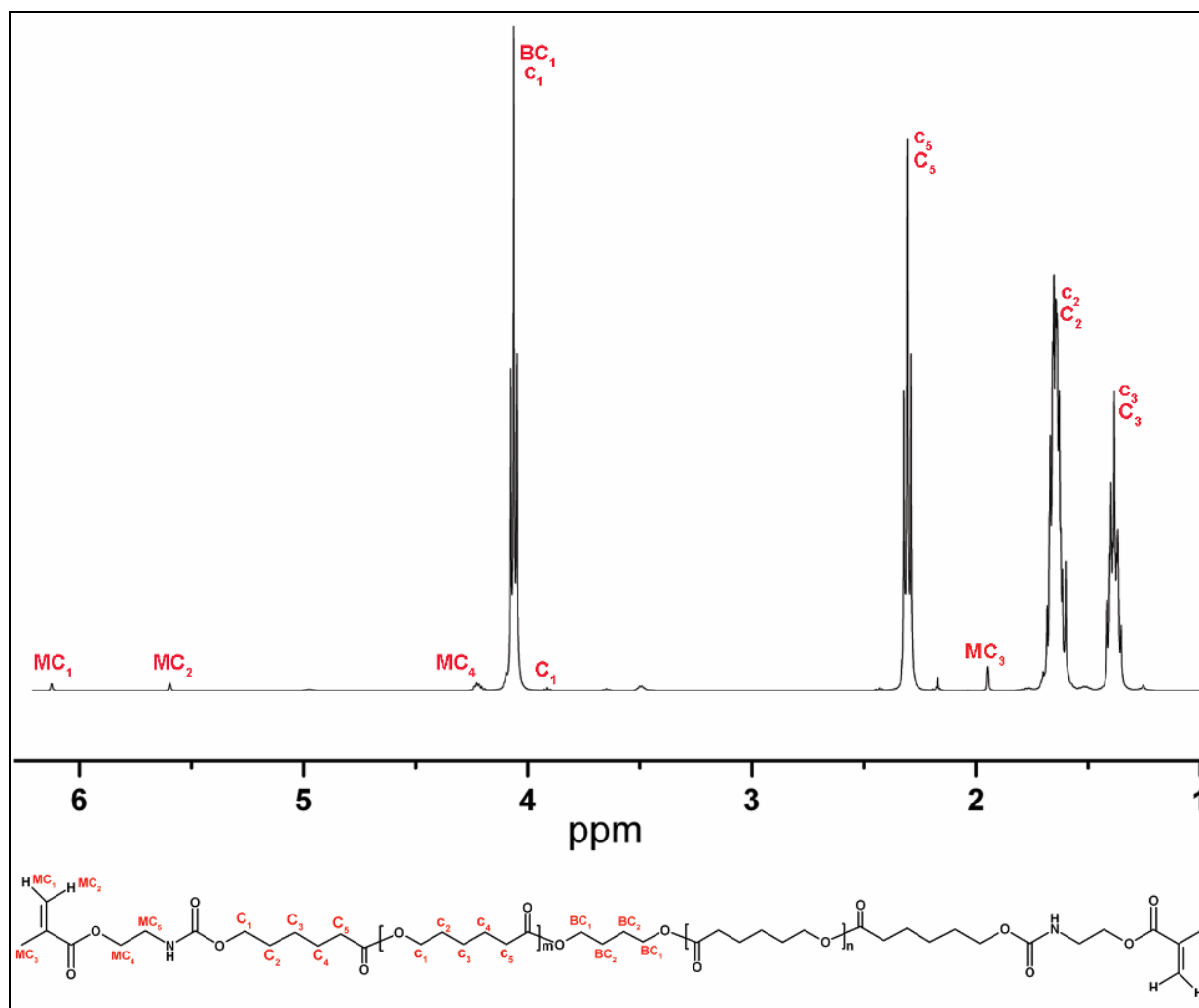
S1: Gel permeation chromatography (GPC)

Multidetector GPC measurements were performed on a system consisting of an isocratic pump 2080 and an automatic injector AS 2050 both (Jasco, Tokyo, Japan), a precolumn, two 300 mm x 8.0 mm linear M columns (Polymer Standards Service GmbH, Mainz, Germany) equipped with two detectors, a RI detector Shodex RI-101 both (Showa Denko Group, Japan) and a dual detector T60A (Viscotek Corp., Houston, US). Chloroform (Acros Organics, Geel, Belgium) with 0.2 vol-% toluene as internal standard was used as eluent at a flow rate of 1 ml·min⁻¹ at 35 °C. Molecular weights and polydispersity (PD) were determined using universal calibration technique based on polystyrene standards with M_n between 1000 g·mol⁻¹ and 3000000 g·mol⁻¹ and using the SEC software WINGPC 6.2 (Polymer Standards Service GmbH, Mainz, Germany).

S2: Nuclear magnetic resonance spectroscopy (NMR)

The degree of methacrylation (D_a) for PCLIDMA was analysed by ¹H-NMR spectroscopy recorded at 25 °C on a 500 MHz Avance spectrometer (Bruker, Karlsruhe, Germany) using deuterated chloroform as solvent and tetramethylsilane (TMS) as internal standard. Experiments were performed at 500 MHz (1H) resonance frequency with the spectral width of 10000 Hz. For determination of degree of methacrylation, the following signals were used: -O-CH₂- (methylene protons from ester group, $\delta = 4.06$ ppm, (C1+BC1)); -CH₂-CO (α -methylene protons from ester group, $\delta = 2.30$ ppm, (C5)), =C-CH₃ (methyl protons in the methacrylate group, $\delta = 1.95$ ppm, MC₃). The D_a for PCLIDMA was calculated according to Eq. (1).

$$D_a = \frac{2}{3} \cdot \frac{I_{M(C3)}}{I_{C1+BC1} + I_{c1} - I_{C5+c5}} \quad \text{Eq. (1)}$$



¹H-NMR spectra of PCLIDMA and nomenclature of the protons of PCLIDMA based on butandiol initiated macrodiols for ¹H-NMR analysis.

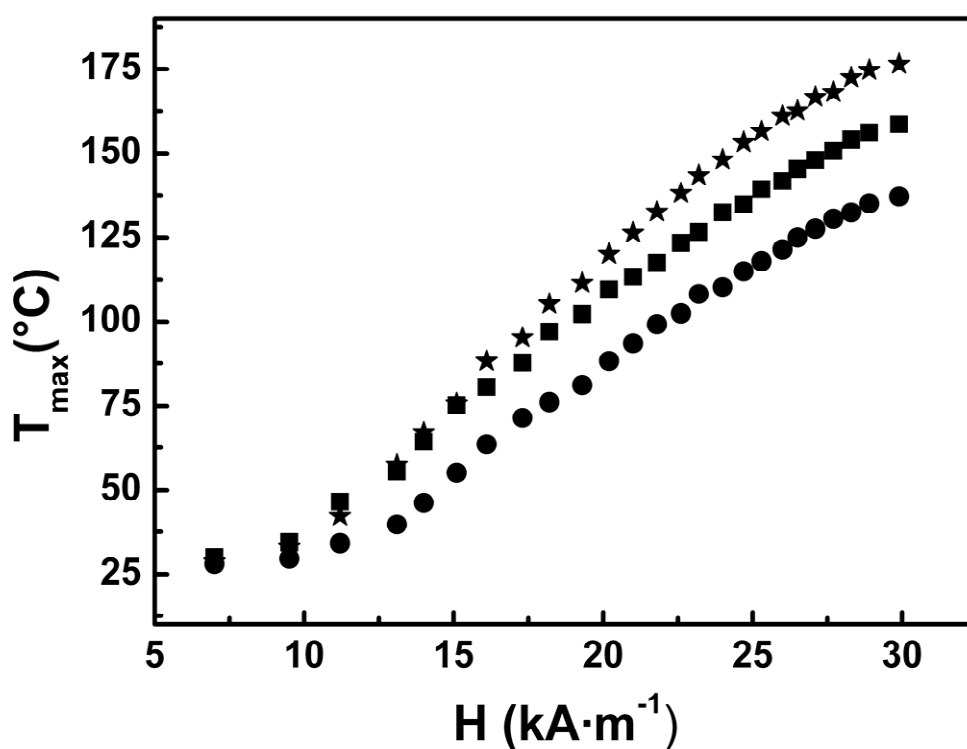
S3: Wide angle X-ray scattering experiments

WAXS measurements were carried out using the X-ray diffraction system Bruker D8 Discover with a two-dimensional detector from Bruker AXS (Karlsruhe, Germany). The X-ray generator was operated at a voltage of 40 kV and a current of 40 mA, producing Cu K α -radiation with a wavelength $\lambda = 0.154$ nm. A parallel, monochromatic X-ray beam was provided by a graphite monochromator and a pinhole collimator with an opening of 0.8 mm. WAXS images were collected at a sample-to-detector distance of 15 cm; samples with a thickness of about 2 mm were illuminated for 120 seconds in transmission geometry. The two-dimensional diffraction images were integrated to obtain plots with intensity versus diffraction angle. These profiles were analyzed using the Bruker software TOPAS to determine the degree of crystallinity (DOC), which is the ratio between

the area of crystalline peaks and the total area below the diffraction curve (area of crystalline peaks plus area of the amorphous halo).

Supporting Figure S1

Achieved maximum temperature (T_{\max}) depending on magnetic field strength (H) for MACL(40) composites with three different filler contents determined by step-wise (ca. $1 \text{ kA}\cdot\text{m}^{-1}$ per step) increase of H from 0 to $29.4 \text{ kA}\cdot\text{m}^{-1}$ at constant frequency $f = 258 \text{ kHz}$. MACL(40)C100, 10 wt-% nanoparticle content (filled circles), MACL(40)C125, 12.5 wt-% nanoparticle content (filled squares), MACL(40)C150, 15 wt-% nanoparticle content (filled stars).



Supporting Video S1 Legend

The real-time thermography video shows the magnetically-induced triple-shape effect of nanocomposite material MACL(40)C125 functionalized with thermomechanical procedures containing two deformation steps TCP-2s-I (top), TCP-2s-II (middle), and a procedure with one deformation step TCP-1s (bottom). The shape changes were recorded while the magnetic field strength was increased to $H = 14.6 \text{ kA}\cdot\text{m}^{-1}$ and $H = 29.4 \text{ kA}\cdot\text{m}^{-1}$ subsequently.

Supporting Table S1: Composition, swelling properties and thermal and mechanical properties of the different MACL polymer networks.

Sample ID ^[a]	G ^[b] [%]	Q ^[b] [%]	DOC ^[c] [%]	X_c ^[d] [%]	$T_g(\text{PCL})$ ^[d] [°C]		$T_{\delta,\max}^{\text{PCL}}$ ^[e] [°C]	$T_m(\text{PCL})$ ^[d] [°C]	$T_{E',\text{inf}}^{\text{PCL}}$ ^[e] [°C]	$T_{\delta,\max}^{\text{PCHMA}}$ ^[e] [°C]	ΔH_m ^[d] [J·g ⁻¹]	(ϵ_B) ^[f] [%]	E ^[f] [MPa]	E' ^[g] (MPa)		
					onset	offset								T_{low}	T_{mid}	T_{high}
MACL(30)	98	888 ± 10	15 ± 1	7	-66	-60	-66	45 ^[d]	50 ^[e]	134	10 ± 2	59 ± 25	124 ± 59	2225	260	3.2
MACL(40)	97	852 ± 10	22 ± 1	13	-66	-58	-66	46 ^[d]	50 ^[e]	134	18 ± 3	226 ± 34	106 ± 60	1942	89	3.5
MACL(50)	97	813 ± 10	31 ± 1	16	-66	-58	-64	47 ^[d]	50 ^[e]	142	22 ± 2	346 ± 42	173 ± 89	1719	15	2.8
MACL(60)	96	797 ± 10	33 ± 3	21	-66	-58	-61	47 ^[d]	46 ^[e]	131	28 ± 3	400 ± 71	90 ± 43	1487	5	2.1
MACL(70)	94	817 ± 10	50 ± 2	27	-65	-58	-63	47 ^[d]	47 ^[e]	-	37 ± 4	477 ± 45	60 ± 26	1225	2	0.2

[a] Sample ID: the two-digit number in brackets gives the weight content of PCLDIMA in the starting material mixture of the organic compounds in wt-% [b] Gel content (G) and swelling degree (Q) determined by swelling experiment in CHCl_3 . [c] Over all degree of crystallinity (DOC) determined by WAXS [d], Data determined by DSC: onset and offset temperature of $T_{g,\text{PCL}}$, $T_{m,\text{PCL}}$, overall melting enthalpy ΔH_m and the crystallinity index X_c . [e] $T_{\delta,\max}^{\text{PCL}}$, $T_{\delta,\max}^{\text{PCHMA}}$ are the transition temperatures determined from $\tan \delta$ -temperature DMTA curve, while $T_{E',\text{inf}}^{\text{PCL}}$ was determined as from the E' -temperature curve. [f] Elongation at break (ϵ_B) and Young's Modulus (E) determined at ambient temperature by tensile test. [g] Storage modulus at $T_{\text{low}} = -10$ °C, $T_{\text{mid}} = 70$ °C and $T_{\text{high}} = 150$ °C determined from the DMTA measurements. The denoted errors are average values.