Supplementary Information (SI) for Green Chemistry.
This journal is © The Royal Society of Chemistry 2026

Supporting Information

Self-Healable Capacitors with Improved Thermostability from Stereocomplex PLA

Neha Mulchandani[1*], Bartosz Mikolaj Gackowski[2], Luciana Tavares[2], William Greenbank[2], Thomas Ebel[2], Anne Ladegaard Skov[1], Anders Egede Daugaard[1*]

[1]*Danish Polymer Center, Department of Chemical and Biochemical Engineering, Søltofts Plads 227, Technical University of Denmark, 2800 Kongens Lyngby, Denmark*

[2]*Centre for Industrial Electronics, University of Southern Denmark, Alsion 2, Sønderborg DK-6400, Denmark*

[*]nehmu@kt.dtu.dk, [*]adt@kt.dtu.dk

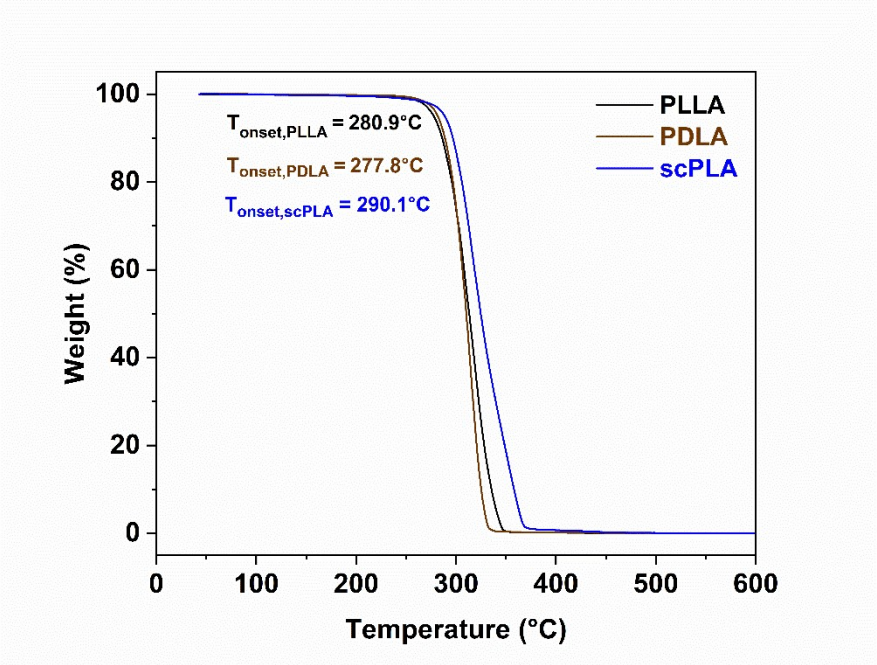
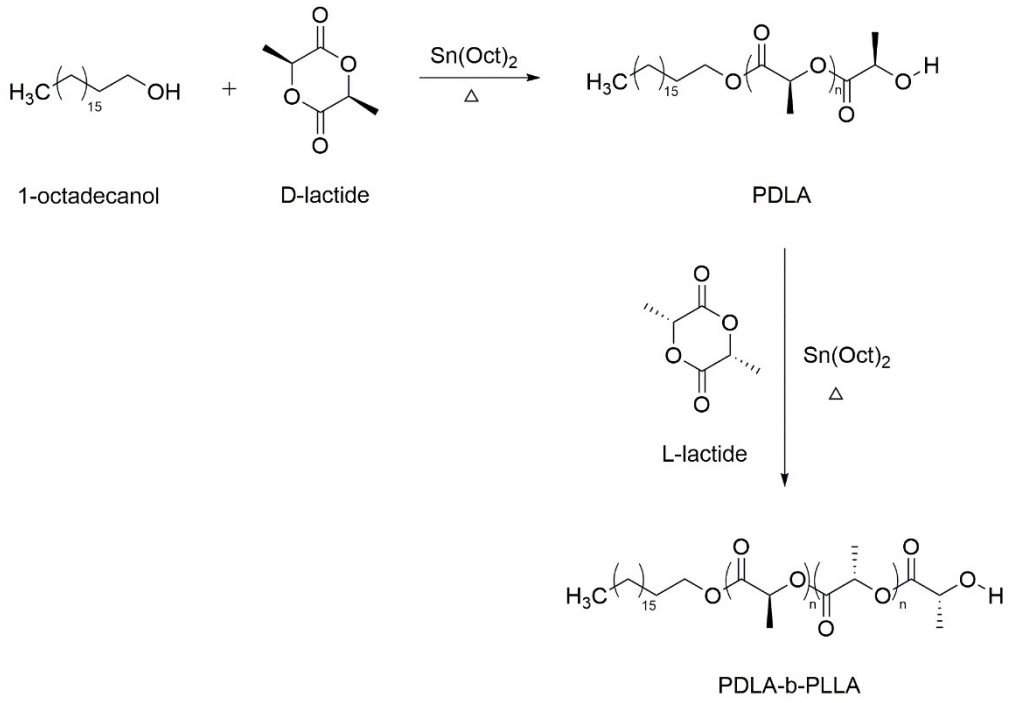


Figure S1: Weight loss (%) as a function of temperature for pure PLLA, PDLA and scPLA.



Scheme S1: Synthesis of stereo-diblock copolymer by two step ROP.

SI-1. Synthesis of stereo-diblock copolymer:

Stereo-diblock copolymer was synthesized by two step ROP as shown in **Scheme S1**. In the first step, mono-hydroxyl terminated PDLA was synthesized by ROP of D-lactide in presence of 1-octadecaol as an initiator and tin octoate catalyst. Briefly, for the case of 10D, the initiator (0.95×10^{-3} mmol) was added to a dried schlenk polymerization tube fixed with a two-way connector and a magnetic stirring bar and evacuated for 1 h with intermittent N_2 purging. This was followed by adding recrystallized D-lactide (6.9×10^{-2} mmol) and evacuated for 6-8 h with intermittent N_2 purging. Finally, a catalyst solution (tin octoate, 0.27×10^{-4} mmol) was added to the apparatus which was then sealed under N_2 atmosphere. The reaction was performed at 180°C for 25 min, and a sample of PDLA was withdrawn for determining the conversion using ^{1}H-NMR. As the conversion of PDLA was higher than 95%, it was subjected to the next step without further purification.

In the second step, the PDLA synthesized as above, was used as a macroinitiator for the synthesis of stereo-diblock copolymer (see **Scheme S2**). Briefly, recrystallized L-lactide (0.14 mmol) was added to the macroinitiator (PDLA, 6.9×10^{-2} mmol) and evacuated at room temperature for 6-8 h. After drying the monomer, the system was filled with N_2 gas and immersed in an oil bath at 190°C to melt PDLA and L-lactide. A catalyst solution (tin octoate,

0.55 × 10^{-4} mmol) was added using a syringe, after the complete solubilization of monomer into the melt of macroinitiator. The reaction was performed at 190°C for 25 min. A sample was withdrawn to determine the conversion of diblock copolymer by ^{1}H-NMR. The synthesized polymer was dissolved in a mixture of dichloromethane + 10% HFIP, followed by precipitating in excess methanol. The diblock copolymer precipitates were filtered and dried in a vacuum oven at 80 °C for 12-16 h. The sample obtained was designated as 'BCP' for further investigations.

SI-2. Fabrication of blends

PLLA was blended with 'DBC' in different ratios to form different blend samples which were regarded as 'PLLA/DBC (20/80)', 'PLLA/DBC (50/50)' and 'PLLA/DBC (80/20)'.

Table S1: Sample characterization

Sample code	$M_{n,th}$ (kDa)	$M_{n,NMR}$ (kDa)	$M_{n,SEC}$ (kDa)	$M_{w,SEC}$ (kDa)	PDI
Homopolymers					
Low MW PLLA		25.4	33.6	59.6	1.77
High MW PLLA	-	98.5	98.5	163.7	1.66
High MW PDLA	-	177.5	73.1	138.2	1.89
Diblock copolymer					
10D (prepolymer)	10.2	10.4	14.9	30.0	2.0
10D-20L (BCP)	30.7	31.8	45.8	83.1	1.8

Note, MW =Molecular Weight

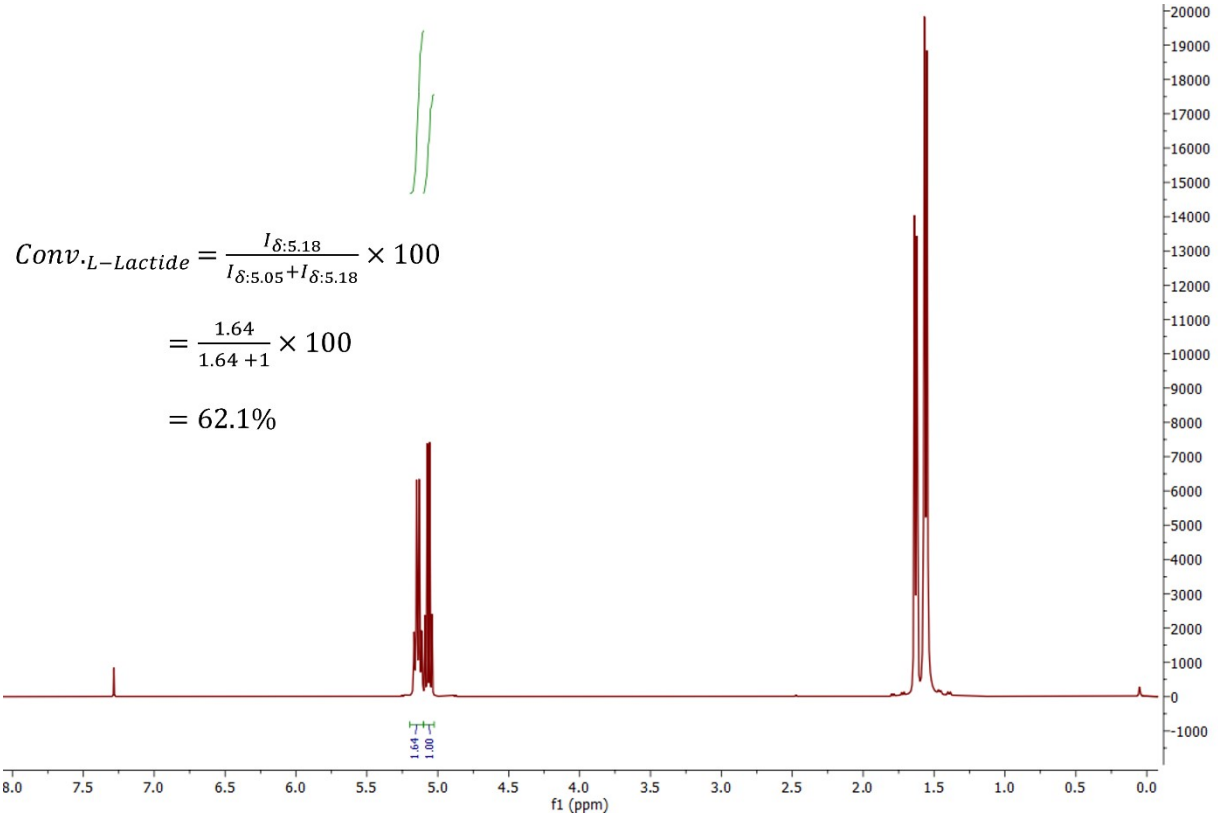


Figure S2: [1]H-NMR spectra of the synthesized high MW PLLA (before purification).

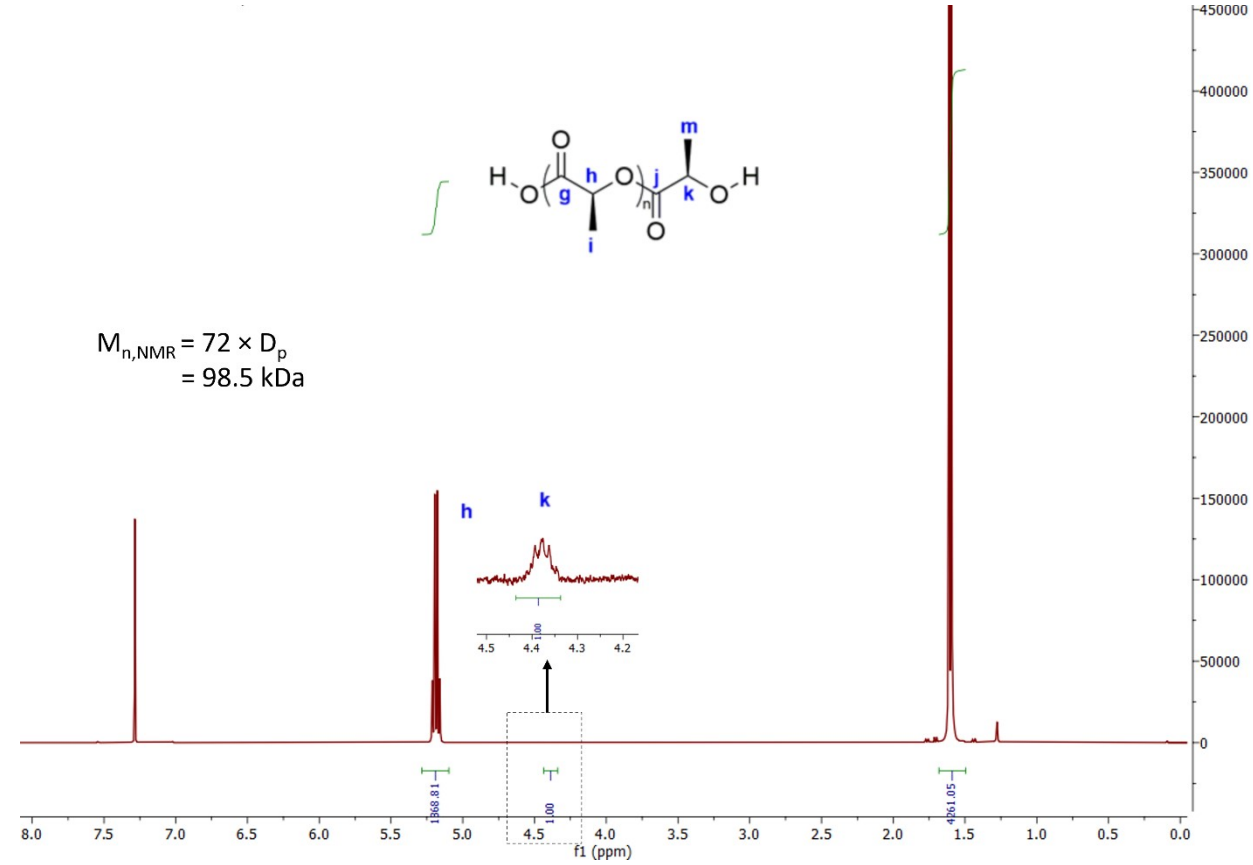


Figure S3: [1]H-NMR spectra of the synthesized high MW PLLA (after purification).

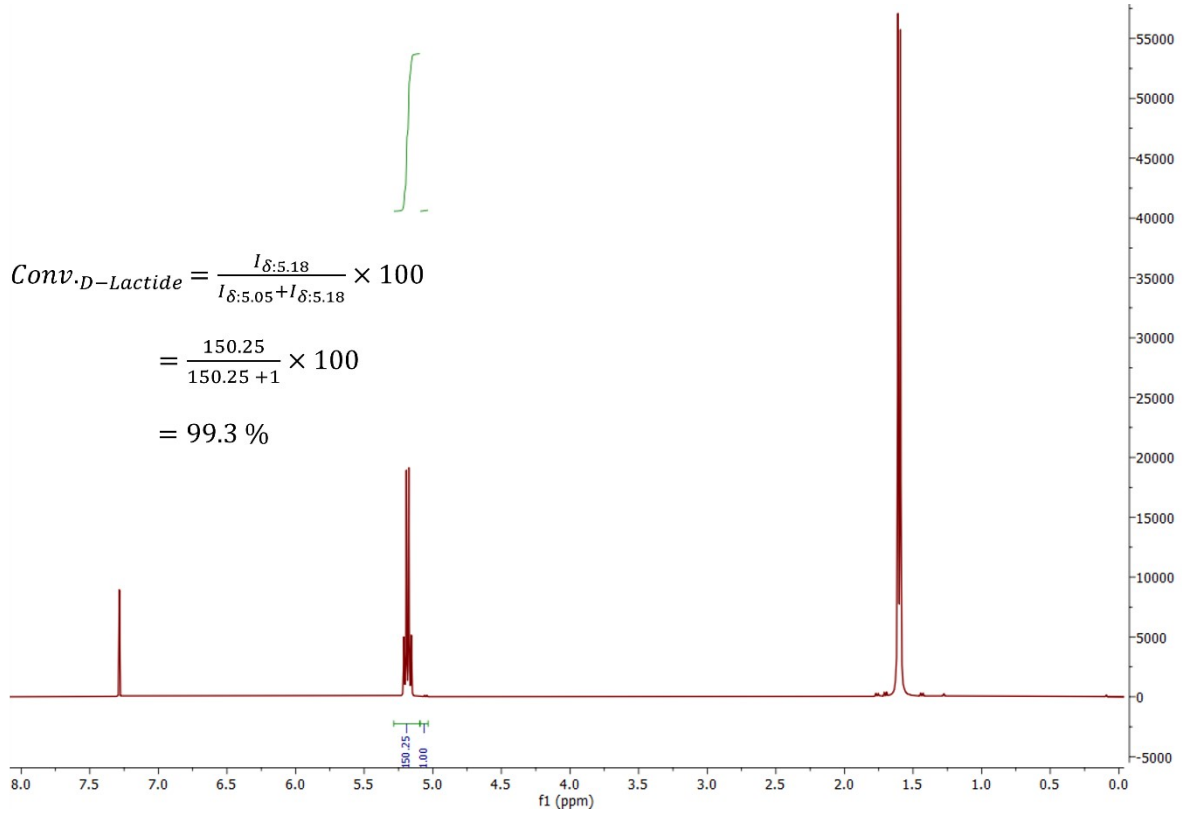


Figure S4: [1]H-NMR spectra of the synthesized high MW PDLA (before purification).

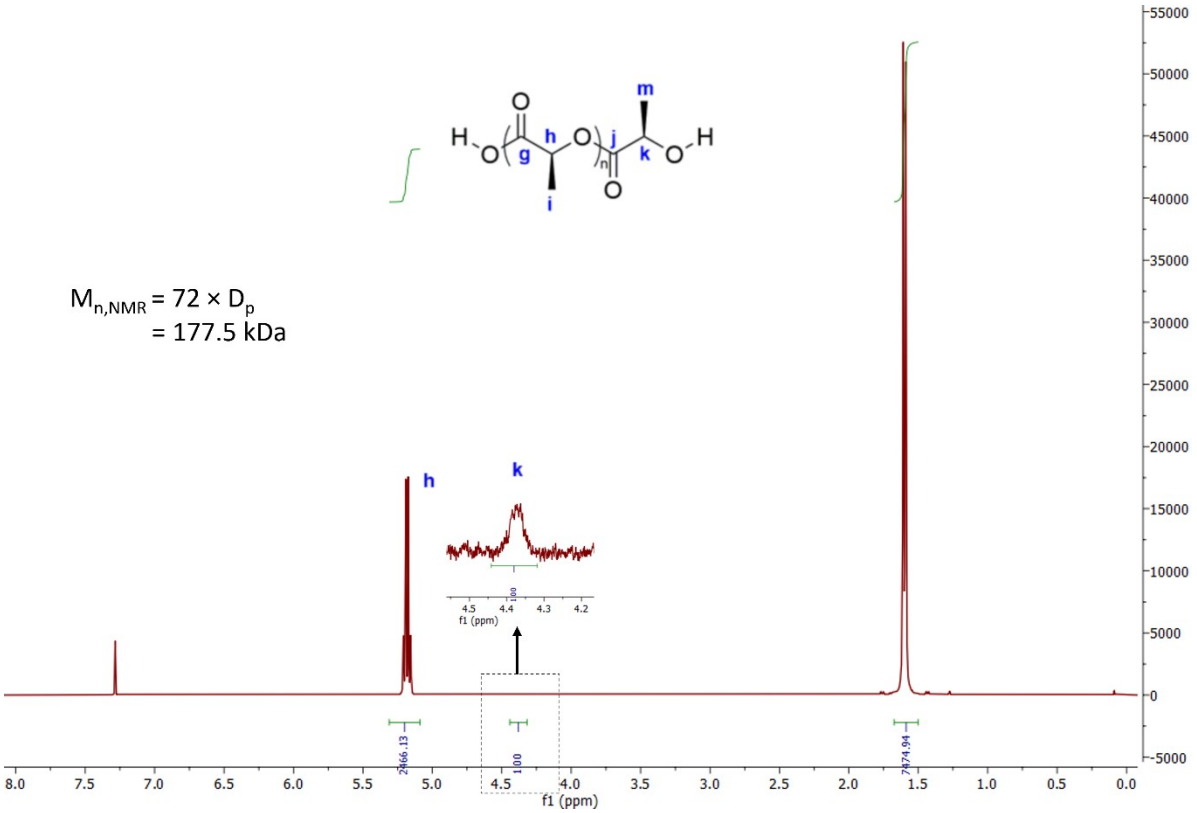


Figure S5: [1]H-NMR spectra of the synthesized high MW PDLA (after purification).

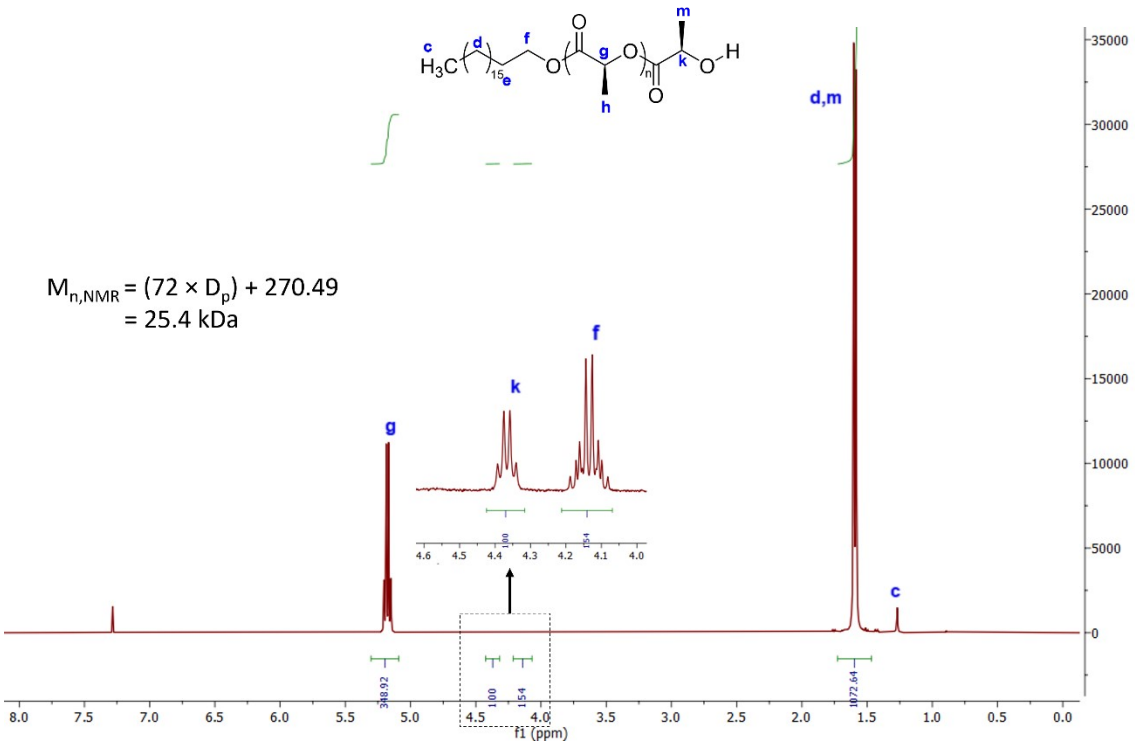


Figure S6: ^{1}H-NMR spectra of the synthesized short chain PLLA (after purification).

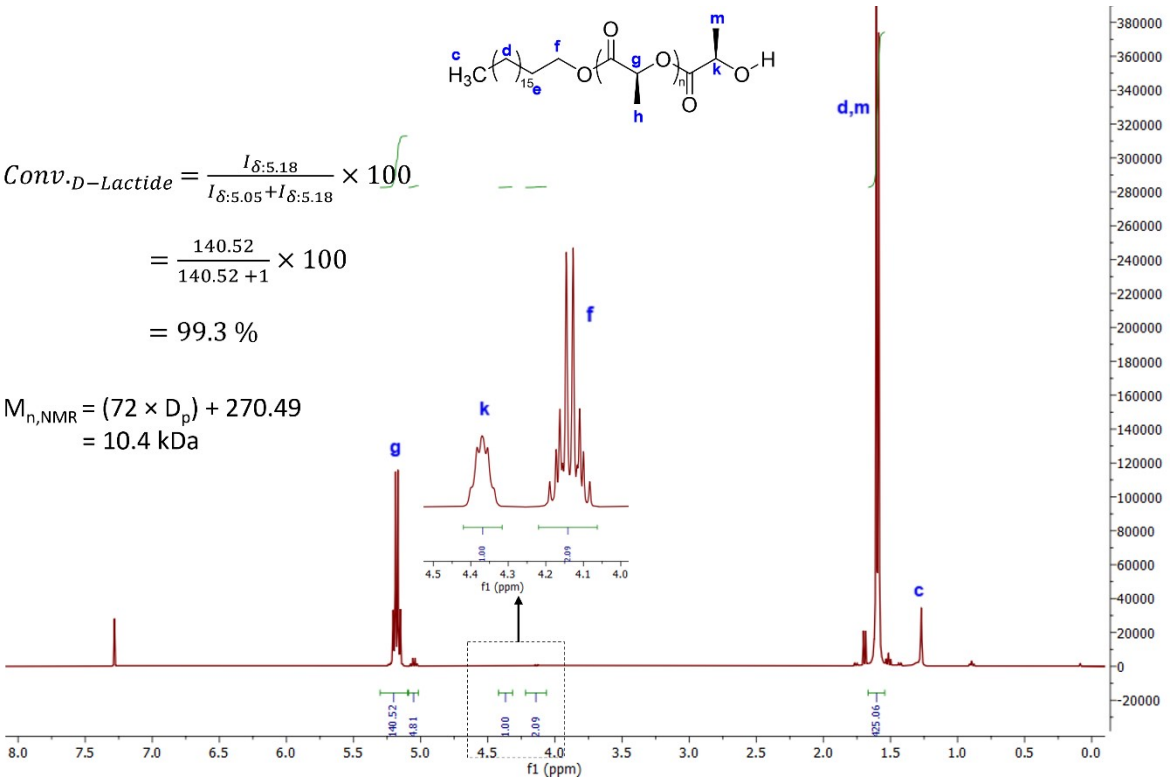


Figure S7: ^{1}H-NMR spectra of the synthesized PDLA (prepolymer).

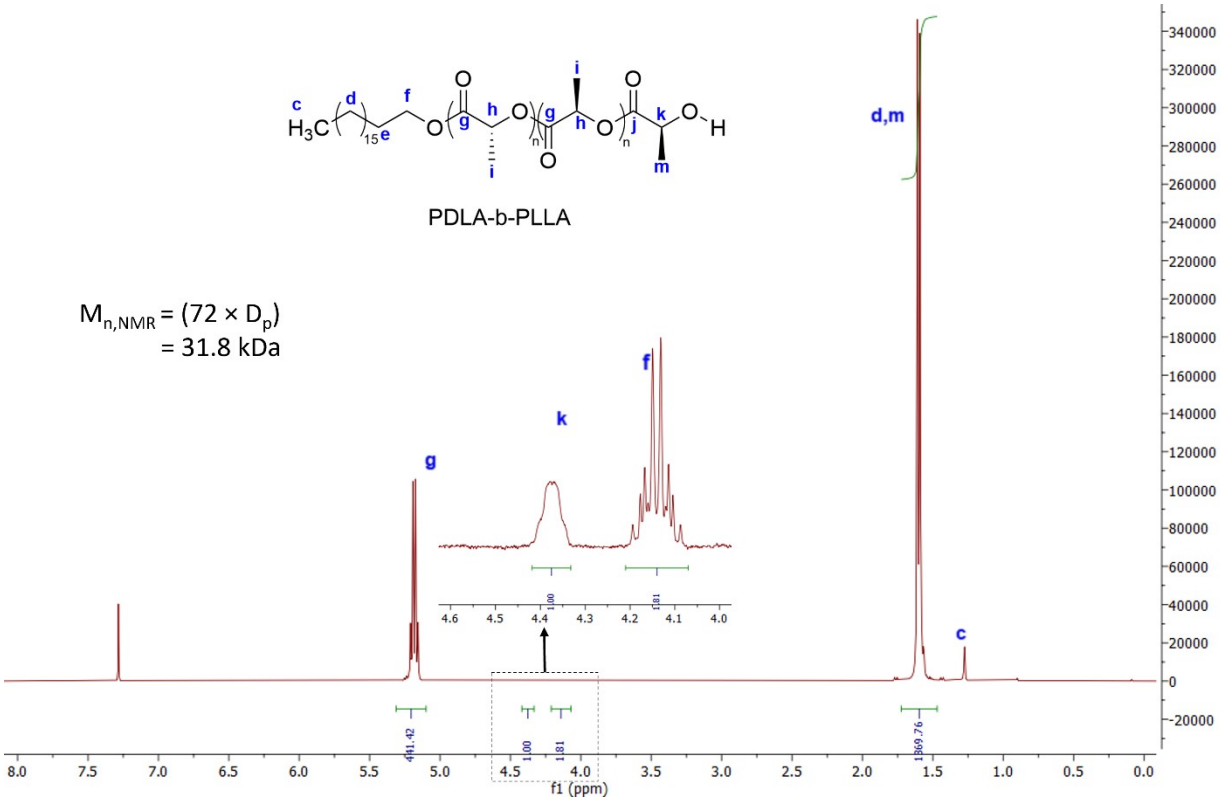


Figure S8: ^{1}H-NMR spectra of the synthesized PDLA-b-PLLA (BCP, after purification).

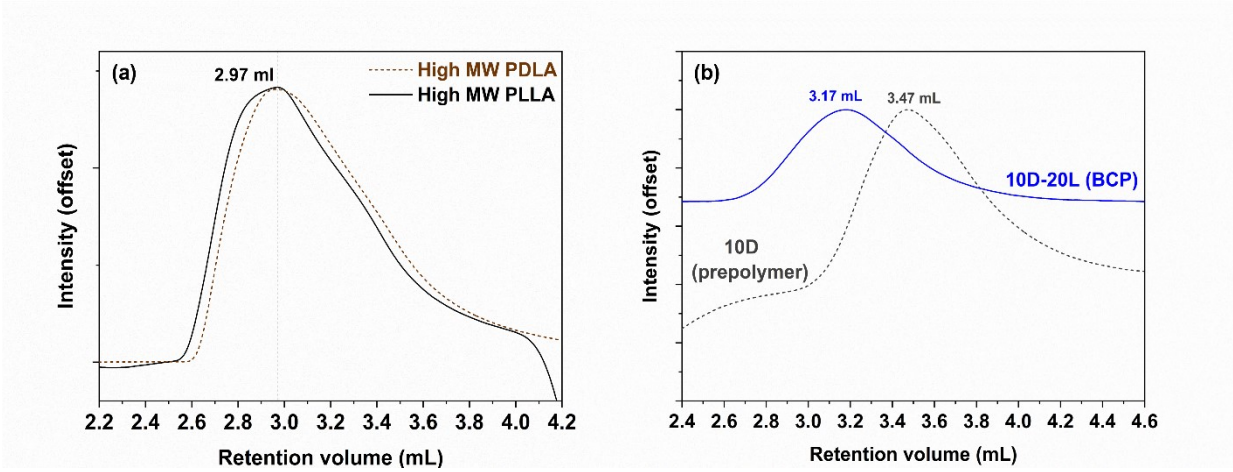


Figure S9: Size exclusion chromatographs of (a) High MW PLLA and PDLA, and (b) 10D (prepolymer), 10D-20L (BCP) synthesized by two-step ROP and high MW PLLA.

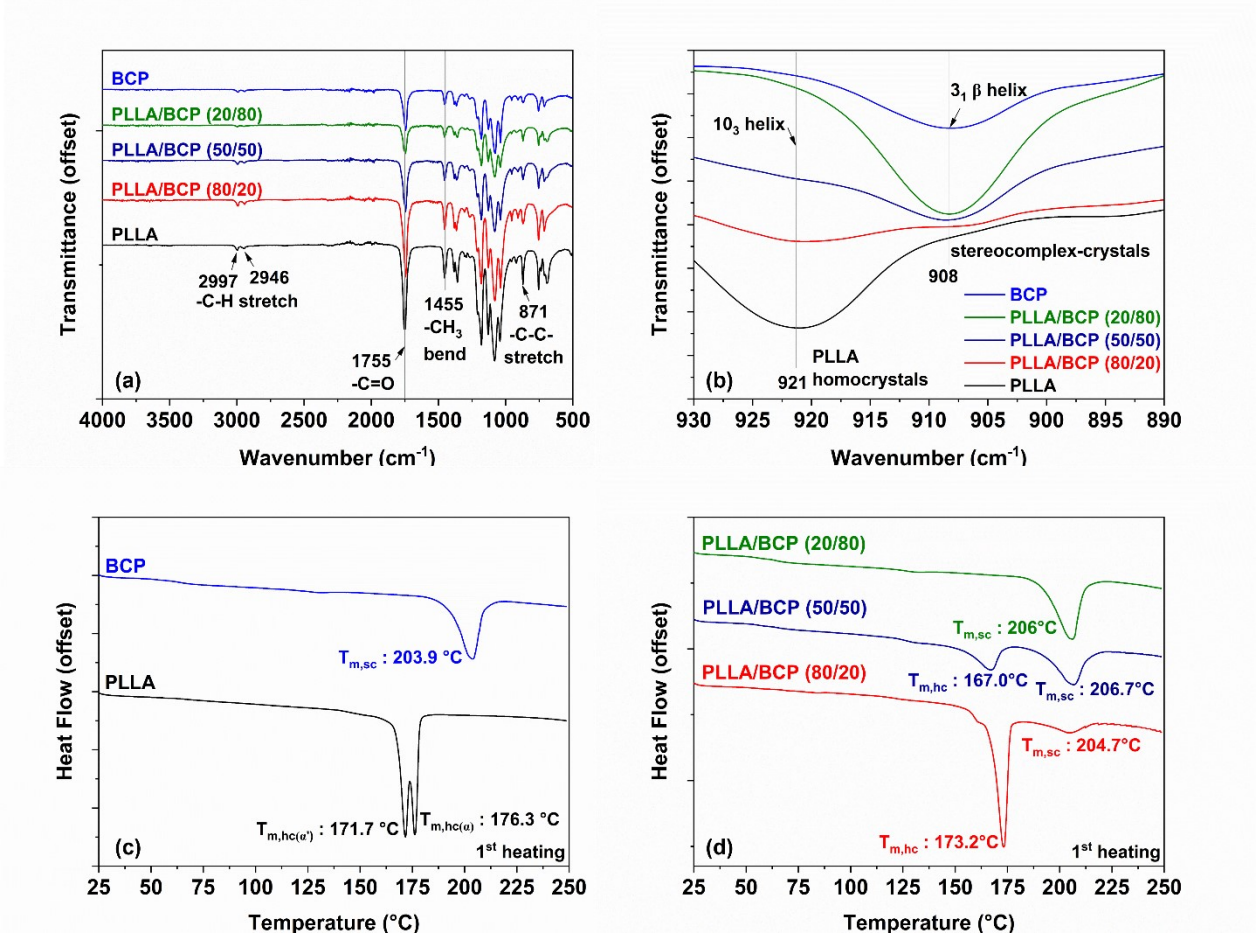


Figure S10: (a) Complete FTIR spectra and (b) magnified FTIR spectra (930 – 890 cm^{-1}) of pure PLLA, BCP, PLLA/BCP (80/20), PLLA/BCP (50/50) and PLLA/BCP (20/80); (c) DSC thermograms of pure PLLA and BCP; (d) DSC thermograms of PLLA/BCP (80/20), PLLA/BCP (50/50) and PLLA/BCP (20/80).

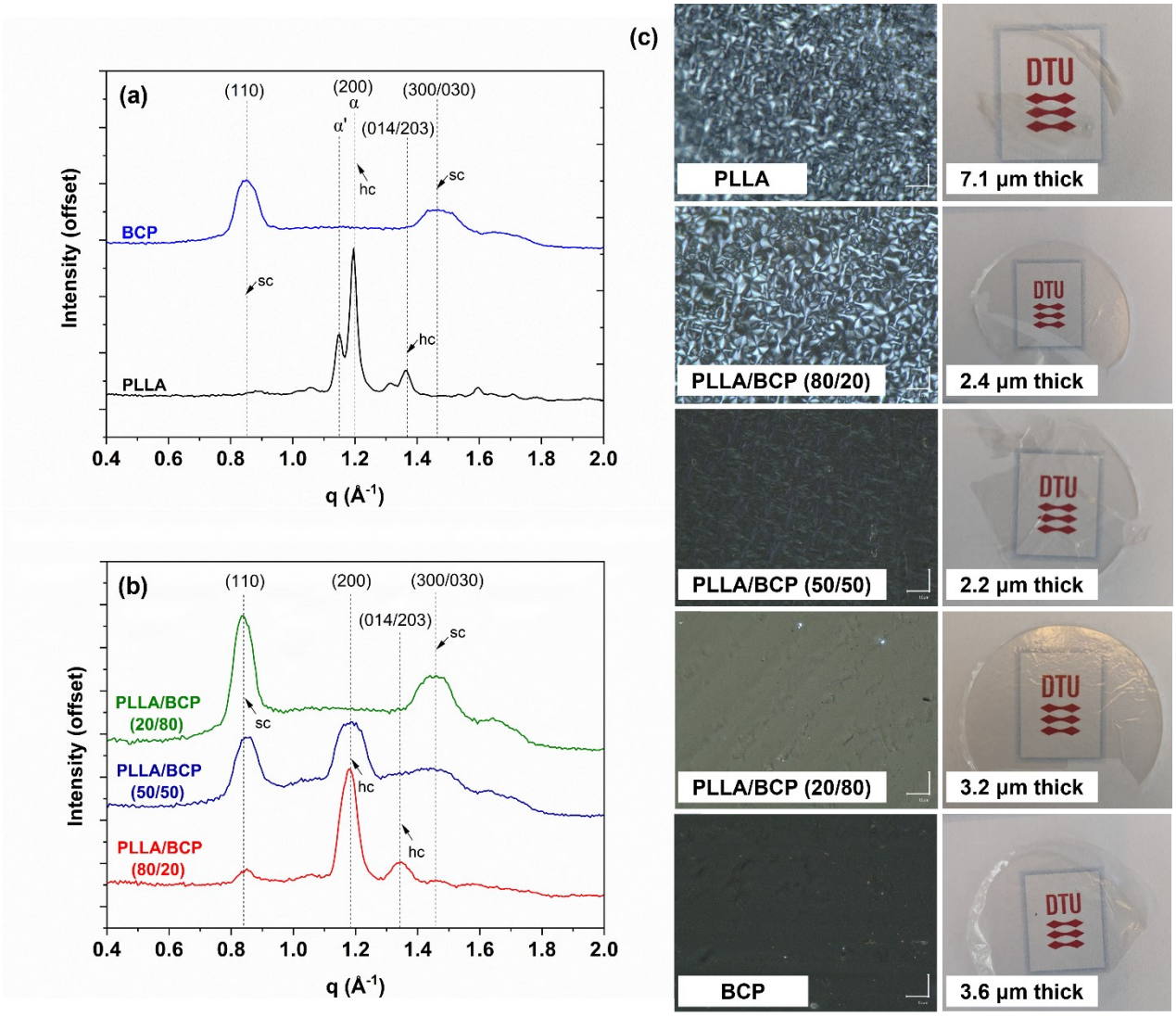


Figure S11: X-ray diffractograms of (a) pure PLLA and BCP, (b) PLLA/BCP (80/20), PLLA/BCP (50/50) and PLLA/BCP (20/80). (c) polarized optical micrographs of PLLA, BCP, PLLA/BCP (80/20), PLLA/BCP (50/50) and PLLA/BCP (20/80) along with their films.

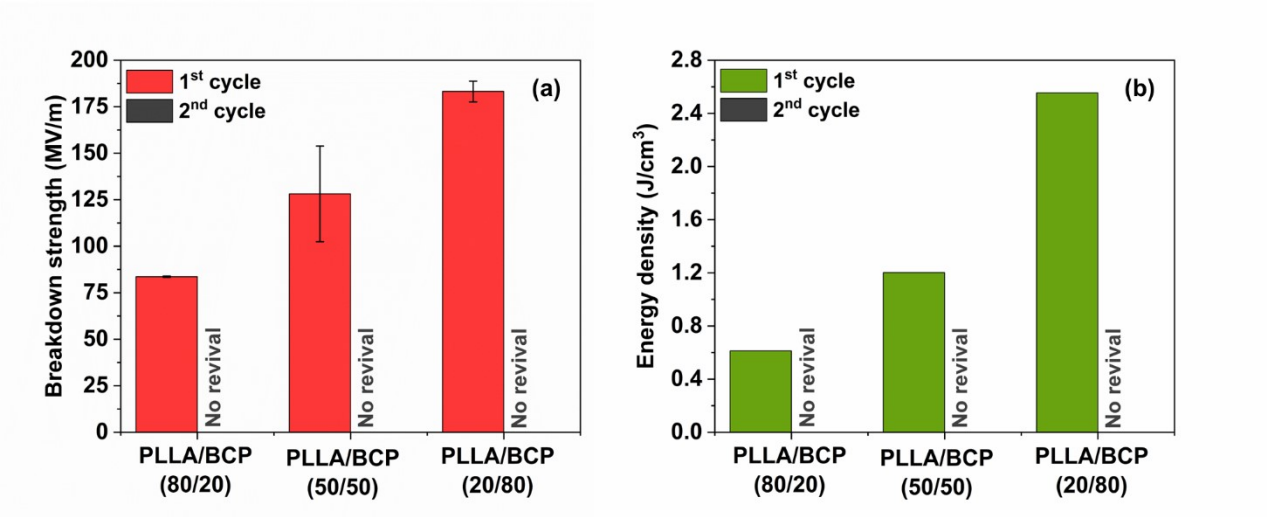


Figure S12: (a) Breakdown strength, and (c) energy density for PLLA/BCP (80/20), PLLA/BCP (50/50) and PLLA/BCP (20/80) after the first and second breakdown cycle.