Polylactide-Based Piezoelectric Blends with Enhanced Thermal Stability and Tunable Mechanical Properties

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● INTRODUCTION ●

Bioresorbable piezoelectric polymeric devices offer great potential for biomedical applications, particularly in implantable bioelectronics, since they can emit electrical signals when activated by physiological dynamic movement or by non-invasive methods such as ultrasound[1].

Polymeric systems derived from **poly(L-lactide) (PLLA)** are a good option as they present piezoelectric response if the macromolecular structure is aligned and subjected to shear stress. The magnitude of the response can be modulated by controlling the degree of **macromolecular orientation** and the degree of **crystallinity**[2].

However, the PLLA presents two major drawbacks[3]:

- Inherent **fragility**.
- Poor **thermal stability.**

To overcome these limitations **poly(L-lactide-co-caprolactone) (PLCL)**, an elastomeric thermoplastic copolymer was used to create **PLLA:PLCL blends** to develop new piezoelectric systems with adjusted mechanical properties, improved thermal stability and shorter bioresorption times[4].

● EXPERIMENTAL PROCEDURE ●

Composition

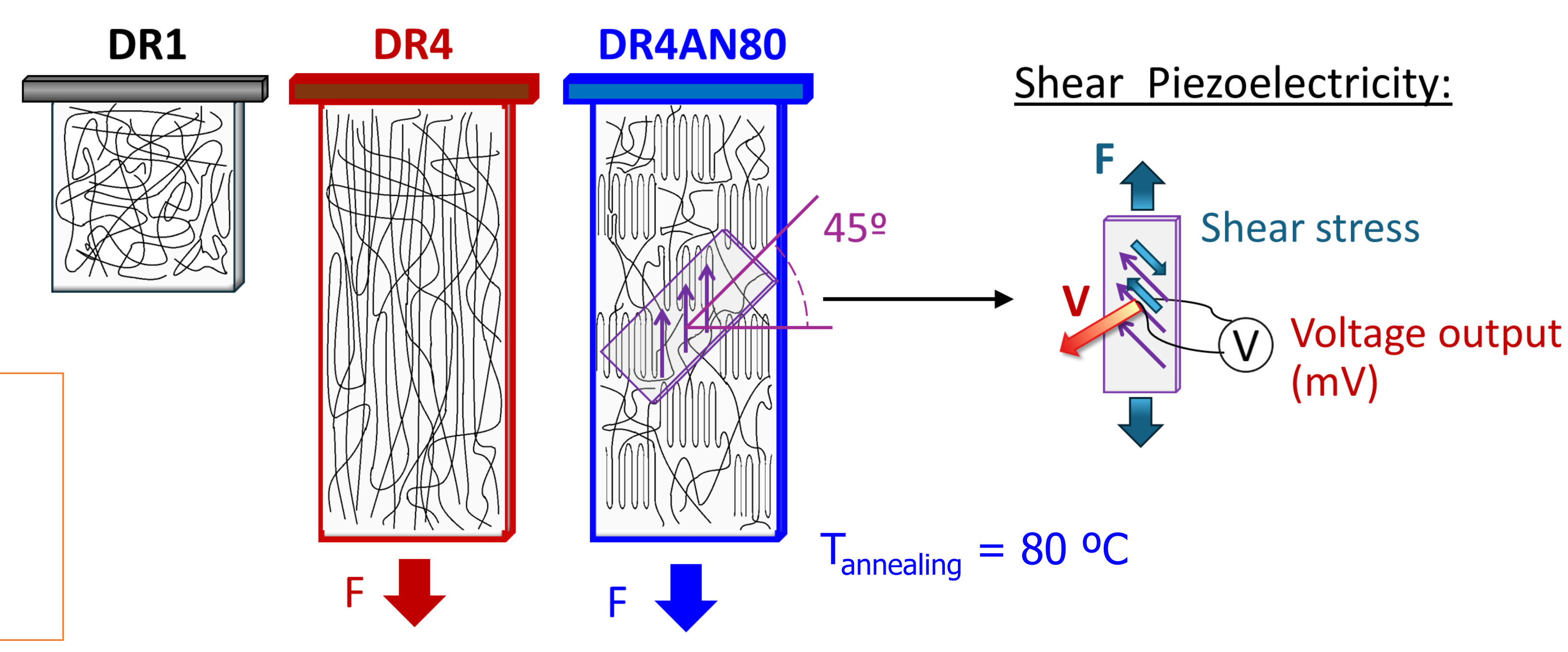
(PLLA:PLCL wt.%):

- PLLA
- 90:10
- 80:20
- 70:30
- 60:40

Bioresorption times

(in PBS at 37 ºC):

- 0 day
- 30 days



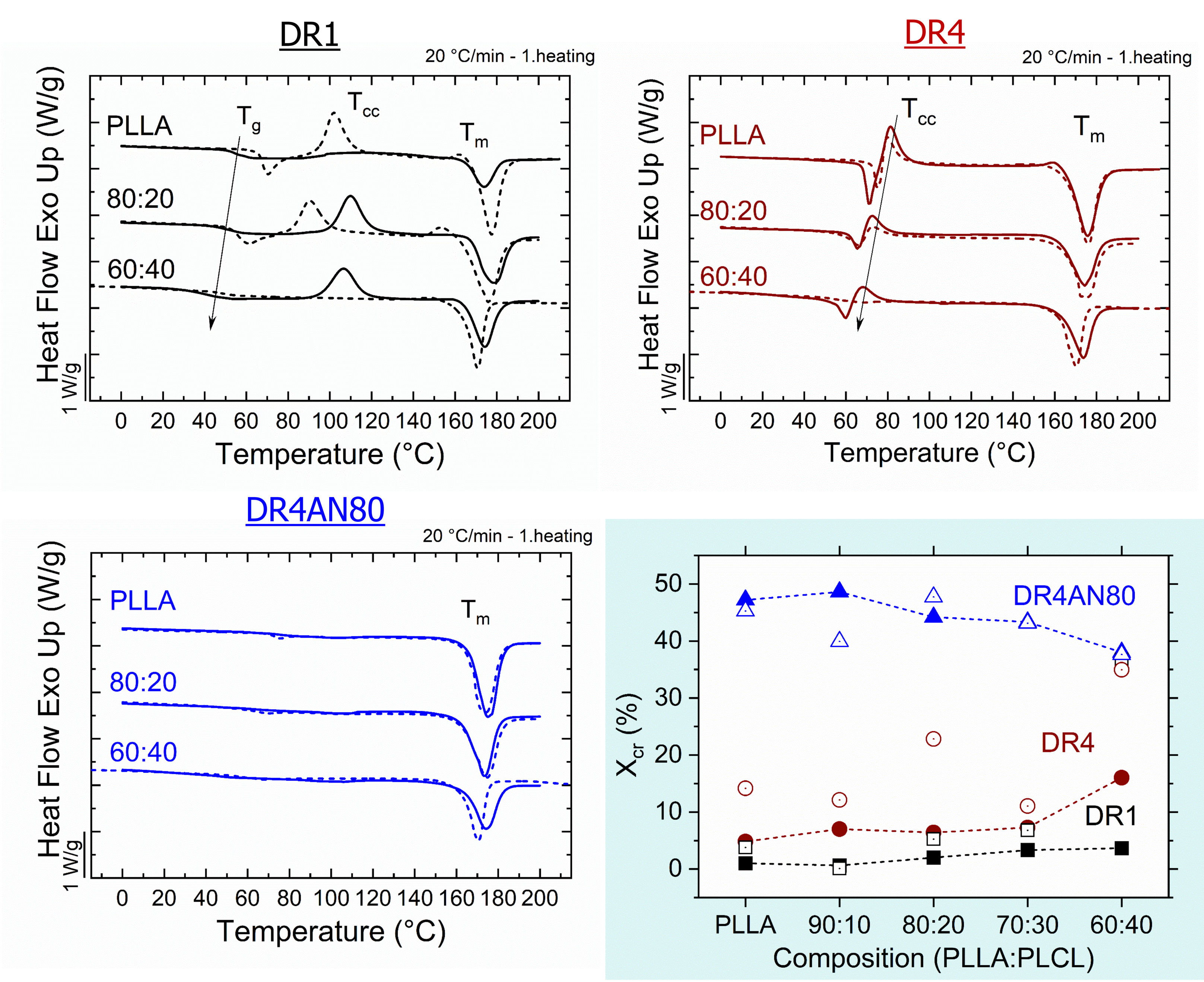
Characterization:

- Differential Scanning Calorimetry (DSC)
- Thermogravimetric Analysis (TGA)
- Tensile Testing
- 2D Wide-Angle X-ray Scattering (2D-WAXS)
- Polarized Raman Spectroscopy (PRS)
- d_{14} Piezoelectric Test

● RESULTS●

Degree of crystallinity

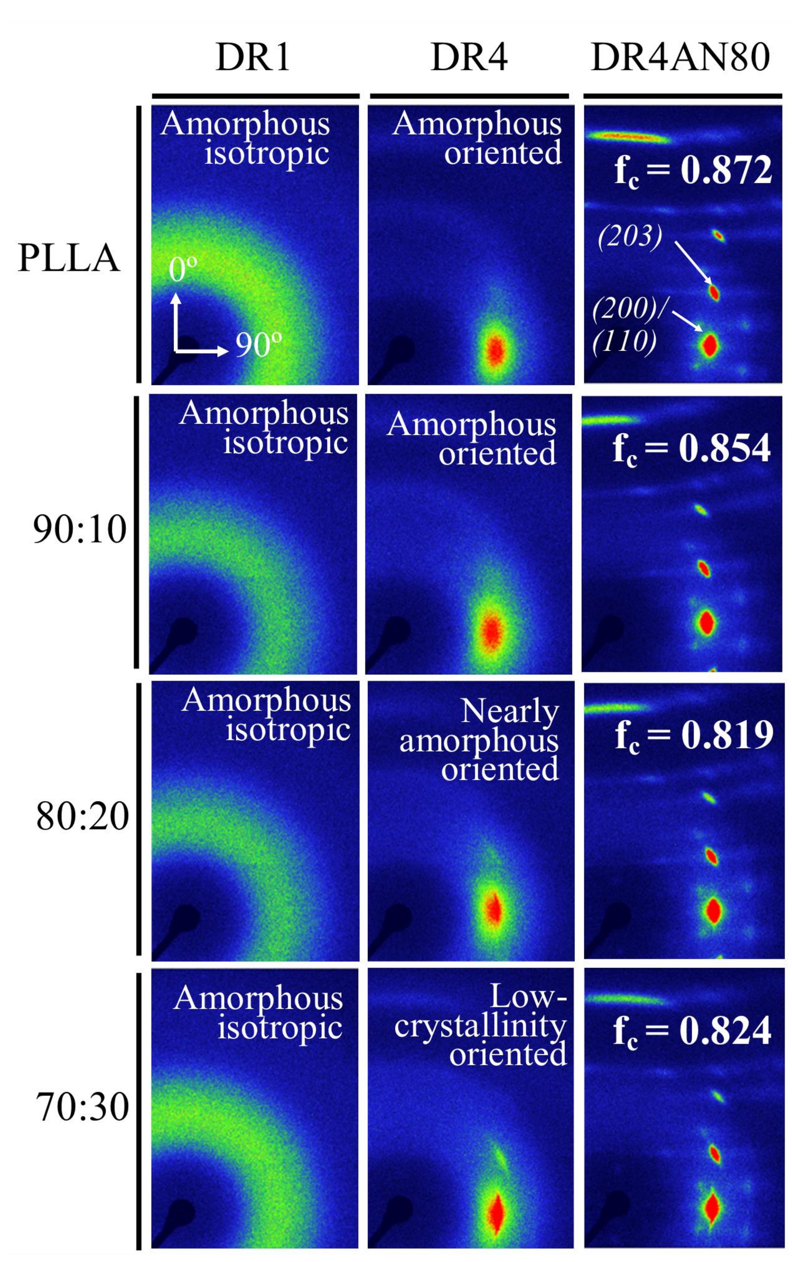
DSC scan



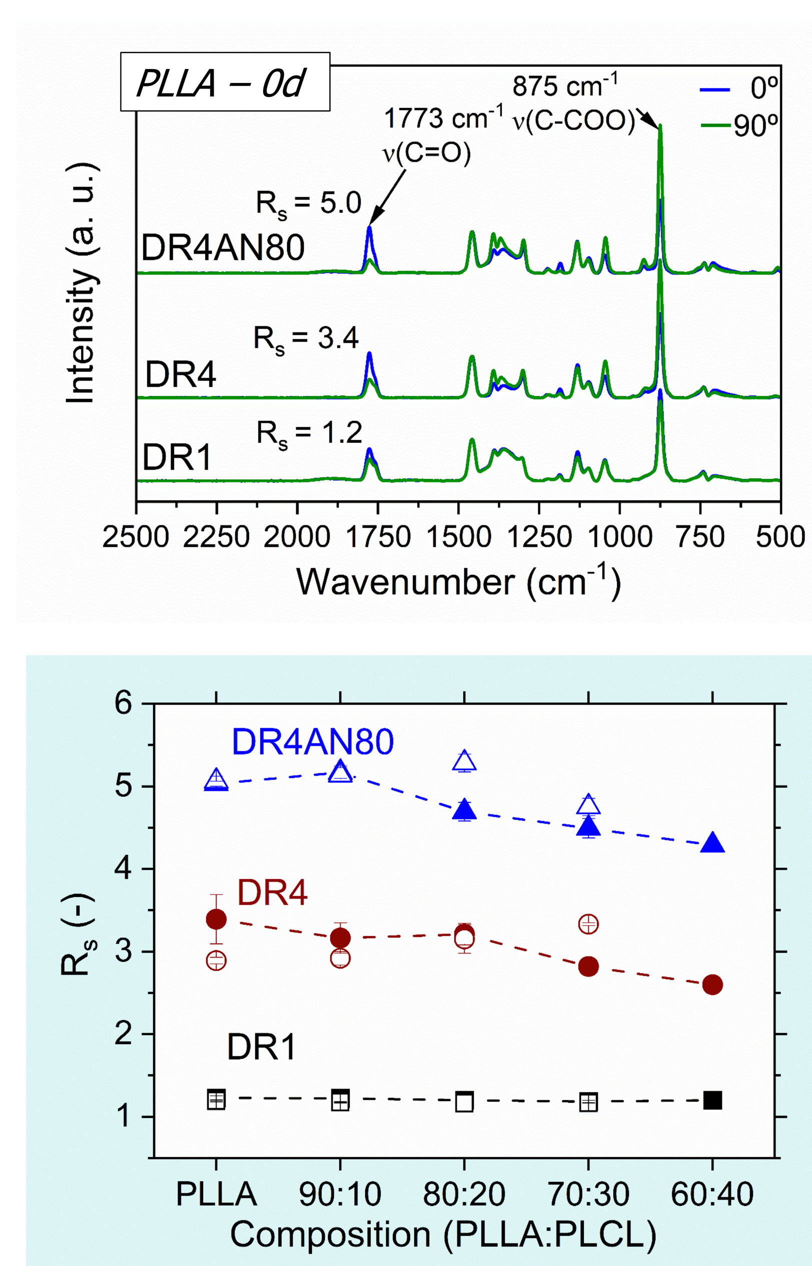
(Solid-symbol: 0d ; Open-symbol: 30d)

Macromolecular Orientation

2D-WAXS analysis



Polarized Raman Spectroscopy (PRS)



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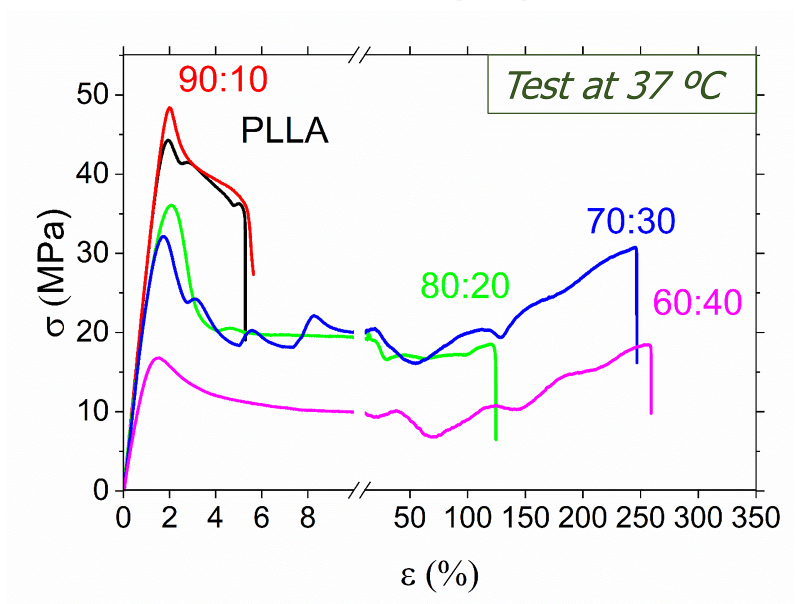
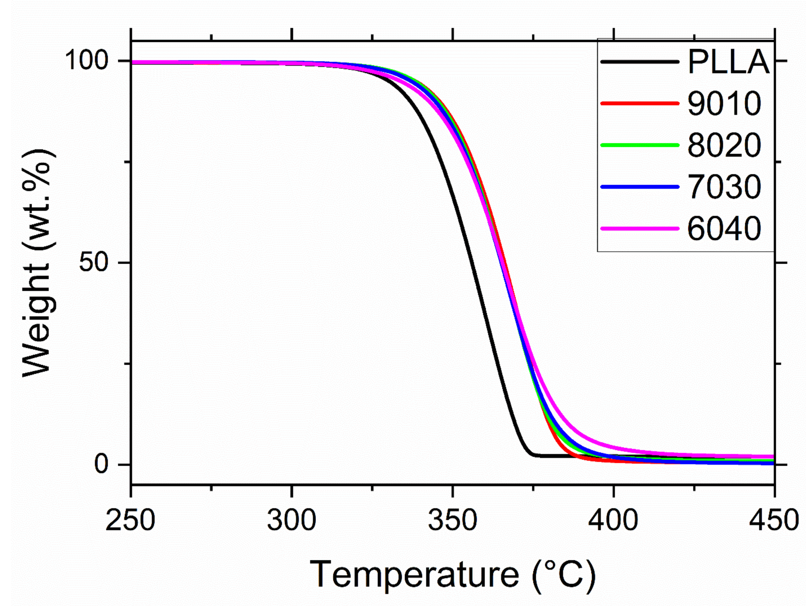
Improved properties

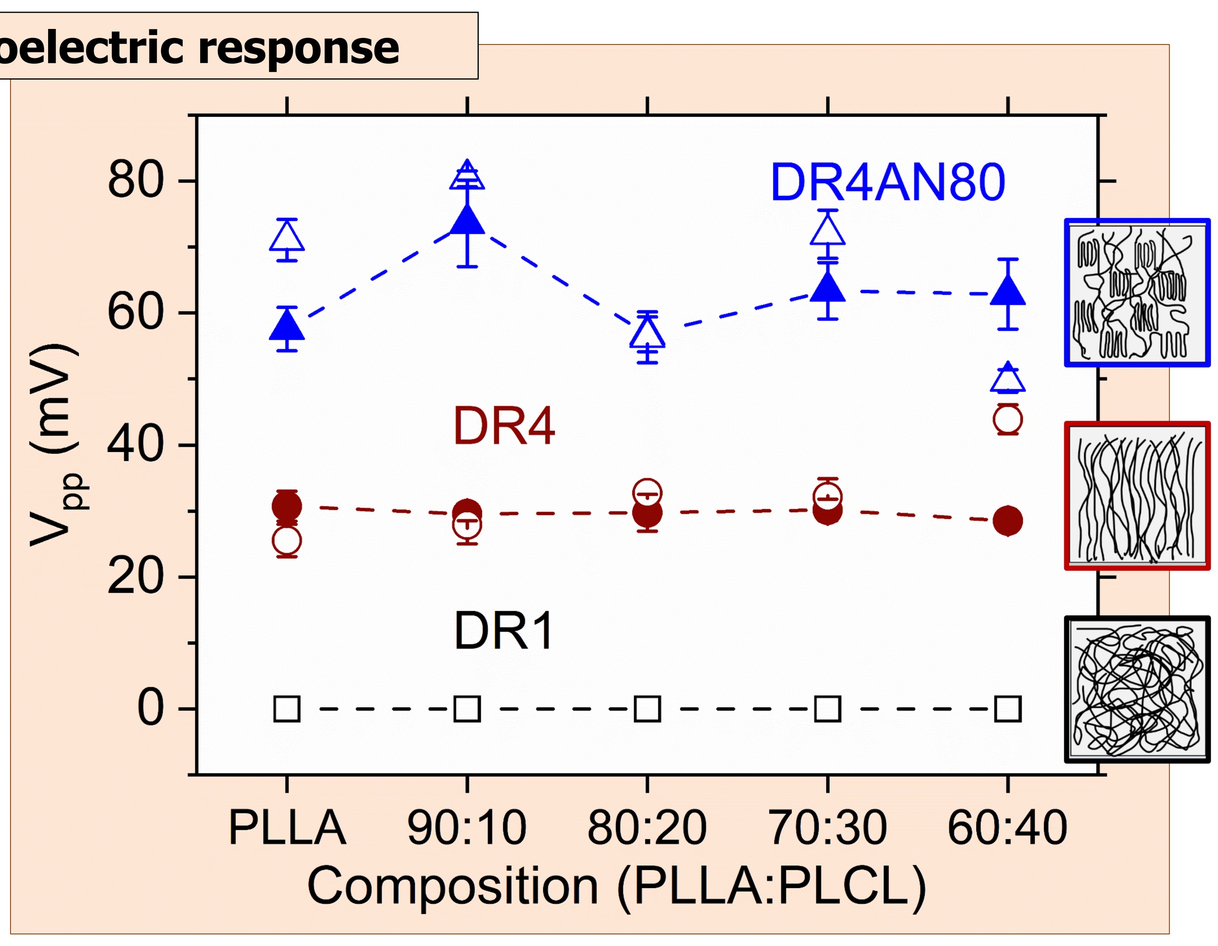
✓ Increased **ductility and toughness** → Modulation of the mechanical properties



✓ Increased **thermal stability** → Possibility to process from the melt



Piezoelectric response



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● CONCLUSIONS●

PLCL can be incorporated up to 40 wt.% without compromising the piezoelectric response while enabling the modulation of mechanical properties. Accordingly, films with reduced stiffness and increased **flexibility and toughness** were obtained. The addition of PLCL also enhanced **thermal stability**, facilitating the processing of the materials via advanced manufacturing techniques (e.g., melt electrowritting or 3D-printing). These findings highlight the potential of PLLA:PLCL piezoelectric blends for applications in implantable bioelectronic devices, soft robotics, and tissue engineering scaffolds, where tunable mechanical and piezoelectric properties are required.

References:
[1] Ribeiro, C., et al. Colloids Surf. B Biointerfaces 136, 46–55 (2015).
[2] Schönlein, R., et al. ACS Appl. Polym. Mater. 6, 7561–7571 (2024).
[3] Polak-Kraśna, K., et al. J. Mech. Behav. Biomed. Mater. 118, 104409 (2021).
[4] Ugartemendia, J. M., et al. Eur. Polym. J. 98, 411–419 (2018).

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