

Understanding Degradation and Mechanical Performance of Hyper-elastic Polylactide Copolymers through Bulk and Ultrathin Film Analysis Correlation

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Introduction

- Degradable polyesters (e.g. polyglycolide, polylactide, and poly(ϵ -caprolactone)) have been extensively used in biomedicine¹
- Understanding their properties is critical for good clinical outcomes
- However**, several aspects are not fully understood especially for novel materials; their molecular processes need to be better understood
- Recently, hyperelasticity was shown in **novel-in house developed system** composed of Poly[(L-lactic acid)-*co*-caprolactone] (PLLAcOCL), blended with Poly(D-lactic acid) (PDLA)²
- Here**, we investigate the degradation behavior of the polymer system (in bulk films, electrospun meshes, and ultrathin films), exploring its applicability as a covering material for cardiovascular implants (i.e. cardiac occluders; example shown in Figure 1)

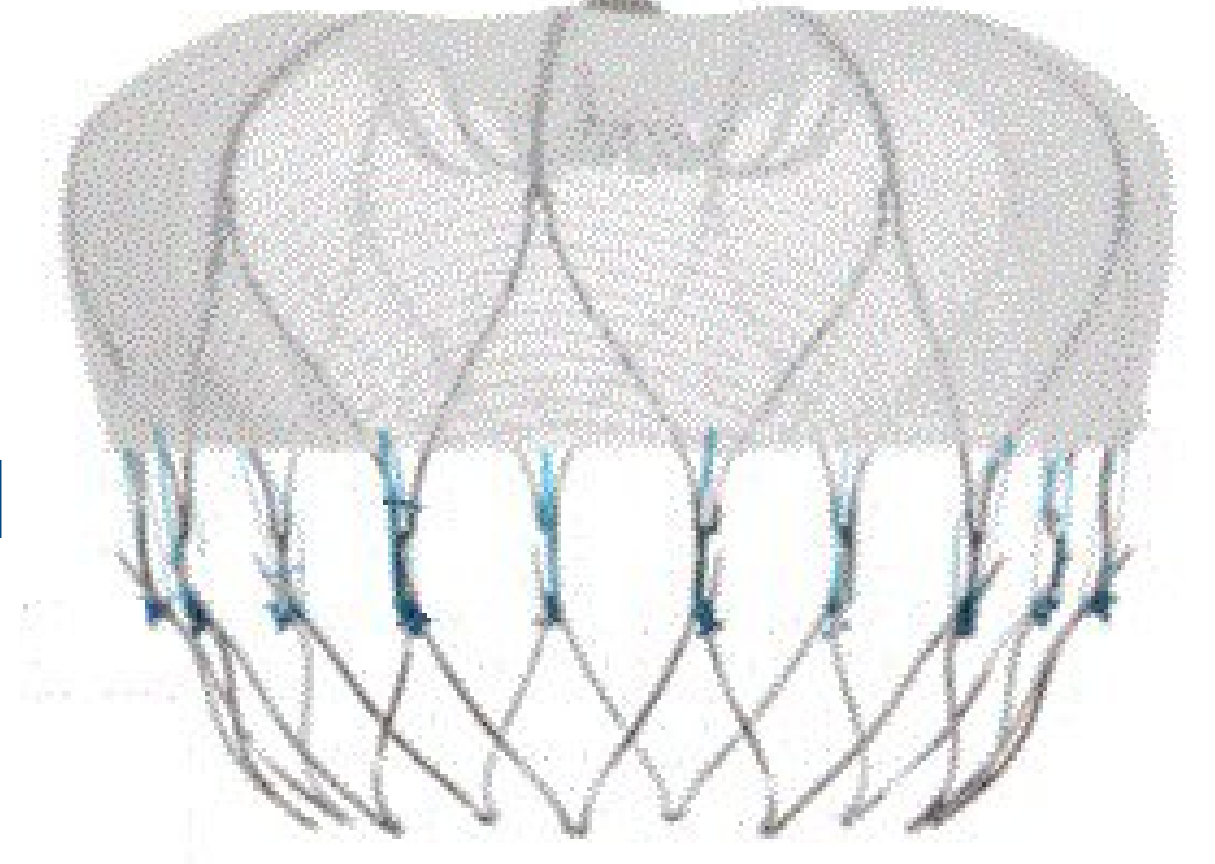


Figure 1: Boston Scientific's Left Atrial Appendage Occluder³

Methodology

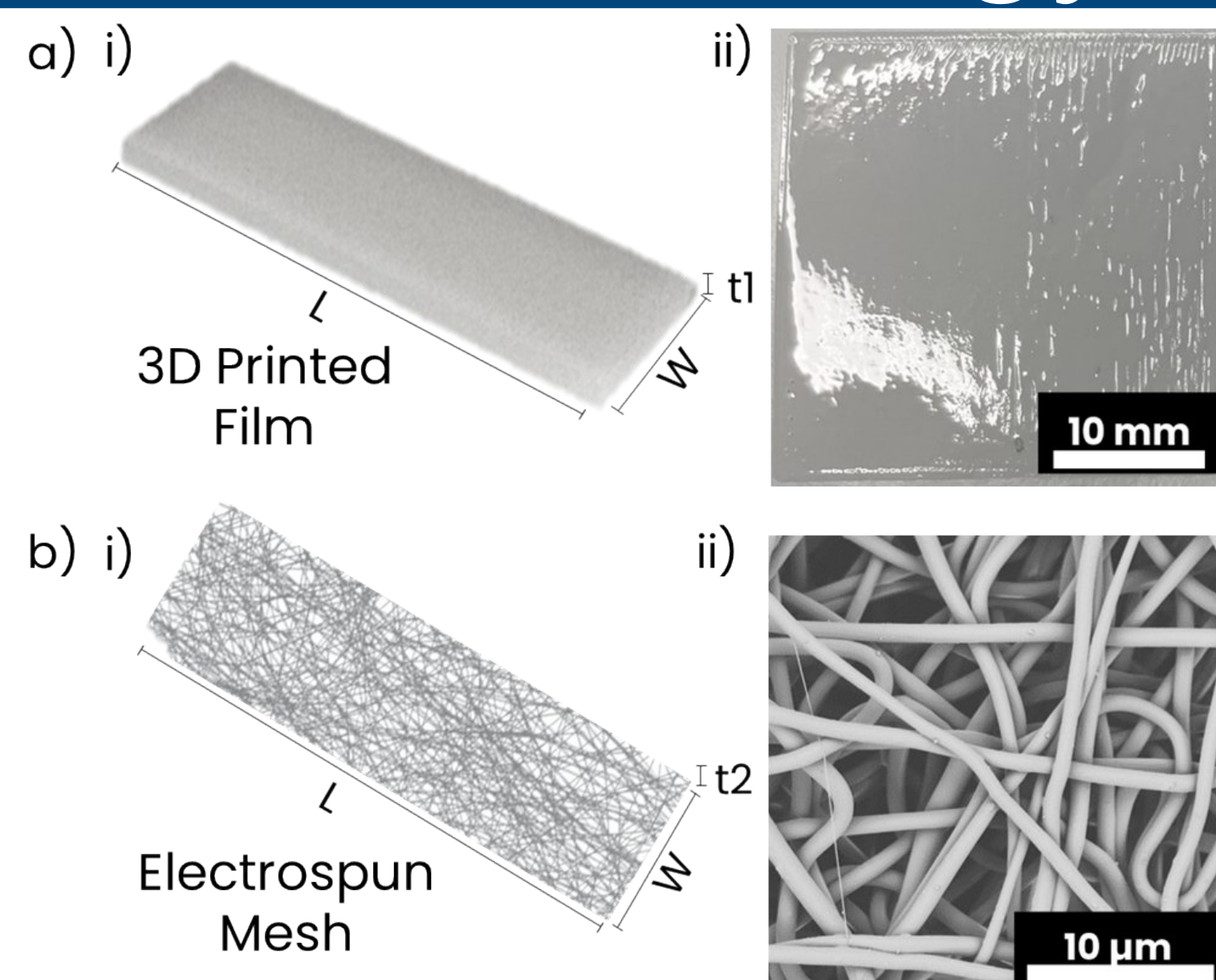


Figure 2: Structure and dimensions of samples prepared for bulk degradation experiments ai) 3D printed film, aii) macroscopic image, bi) electrospun mesh, iv) SEM image prepared by blending 95:5 PLLAcOCL203:PDLA20. $L=30\text{mm}$, $W=10\text{mm}$, $t1=5\text{mm}$, $t2=0.5\text{mm}$

Degradation Conditions: in PBS, 37 °C

Results

➤ Increase in crystallinity during degradation leads to changes in mechanical properties

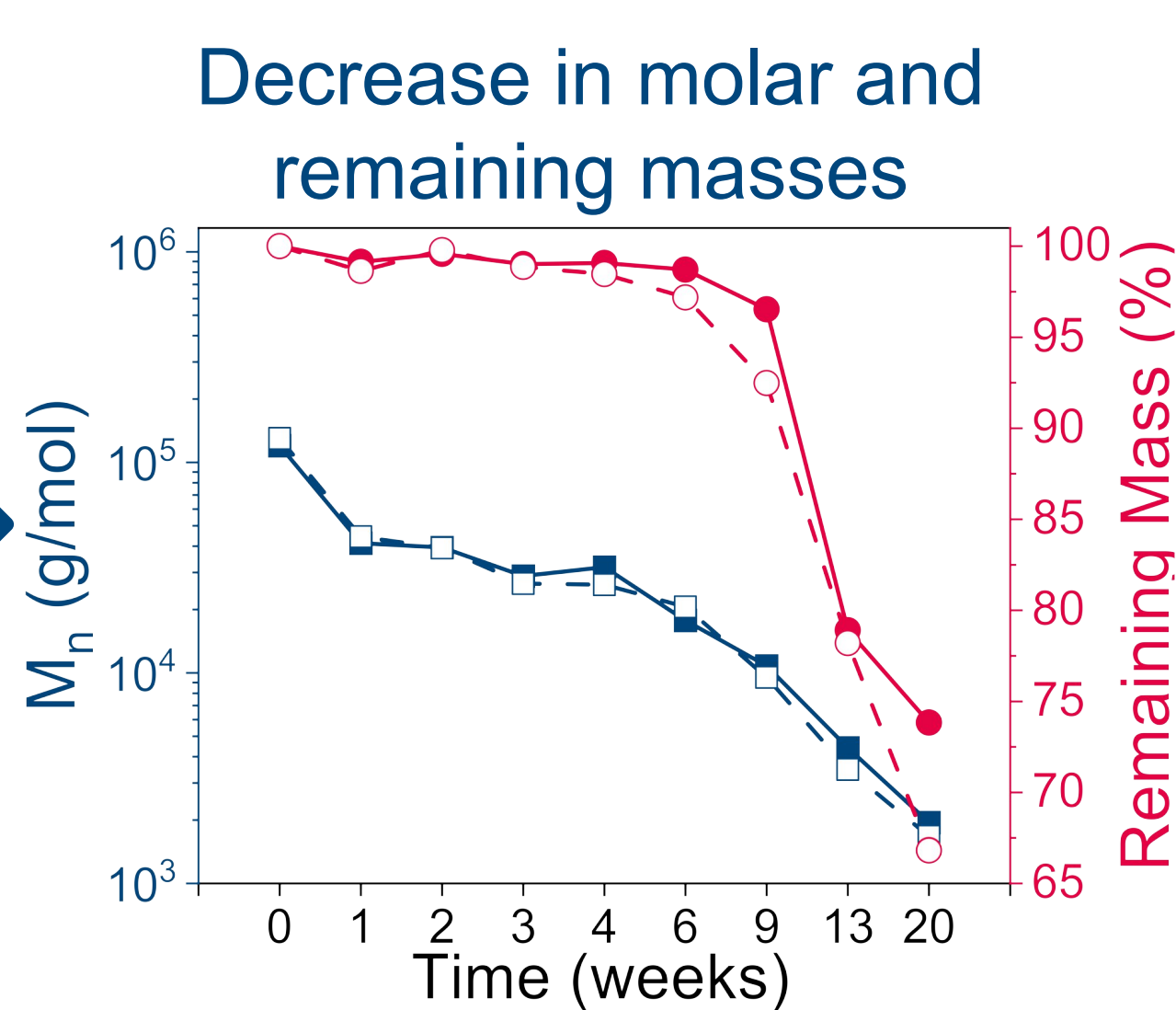


Figure 3: Reduction of both Molar (black) and remaining mass (red) over 20 weeks

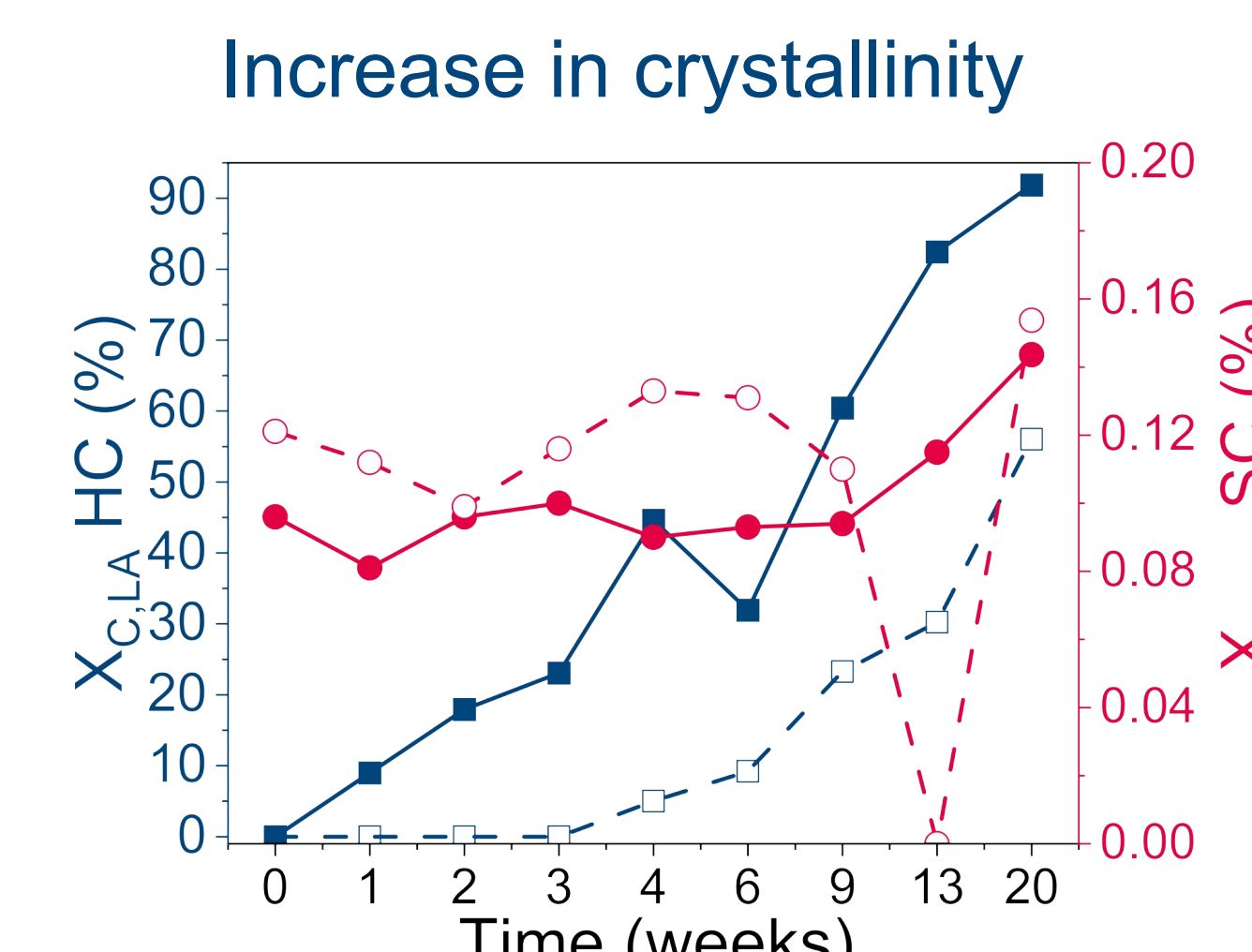


Figure 4: Increase in relative crystallinity of homocrystallites (HC) and stereocrystallites (SC) over time due to the increased movement of chains

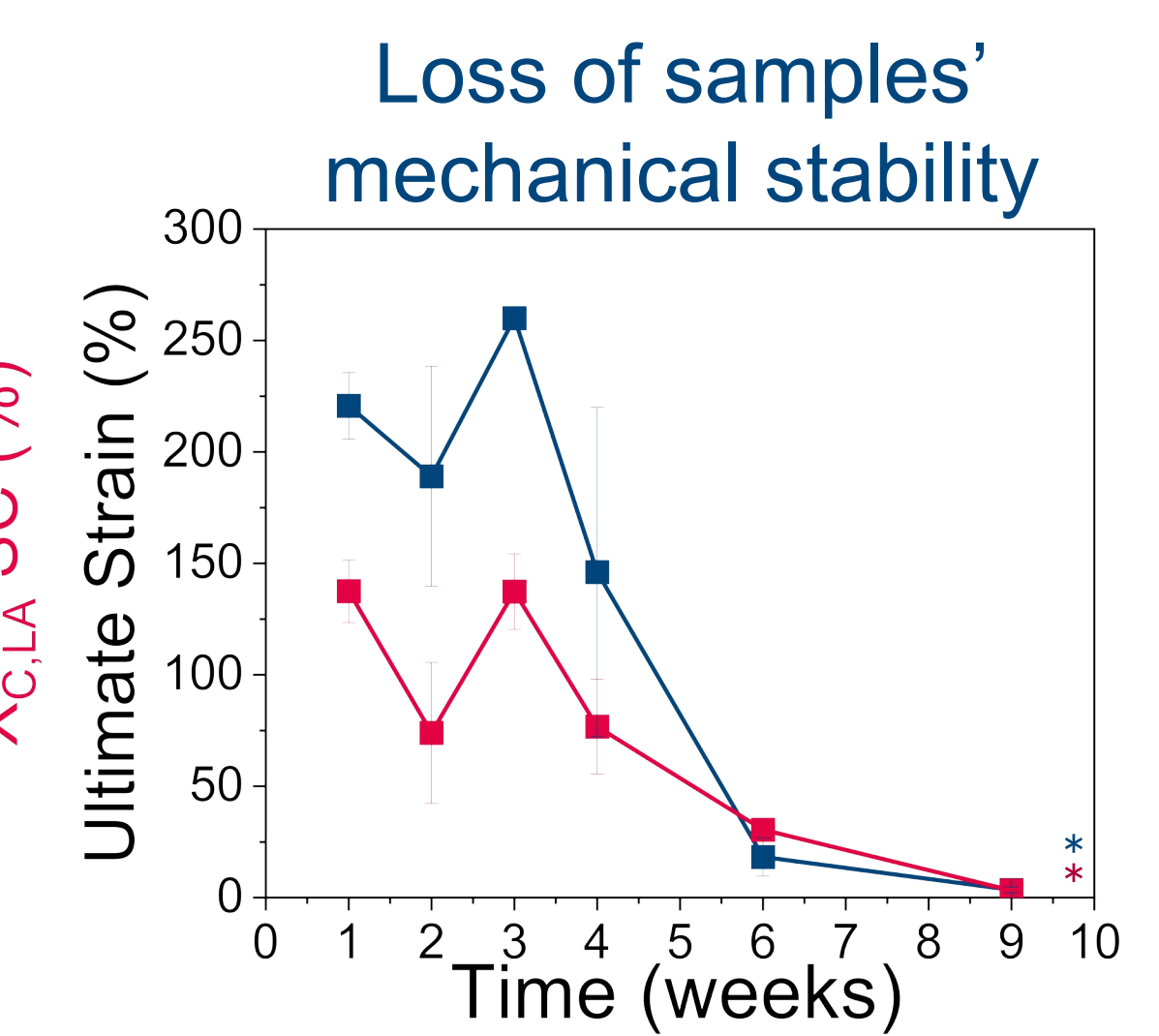


Figure 5: Decrease in ultimate strain over time

Bulk Degradation

Ultrathin Films Degradation

➤ Via the Langmuir Technique^{4,5}

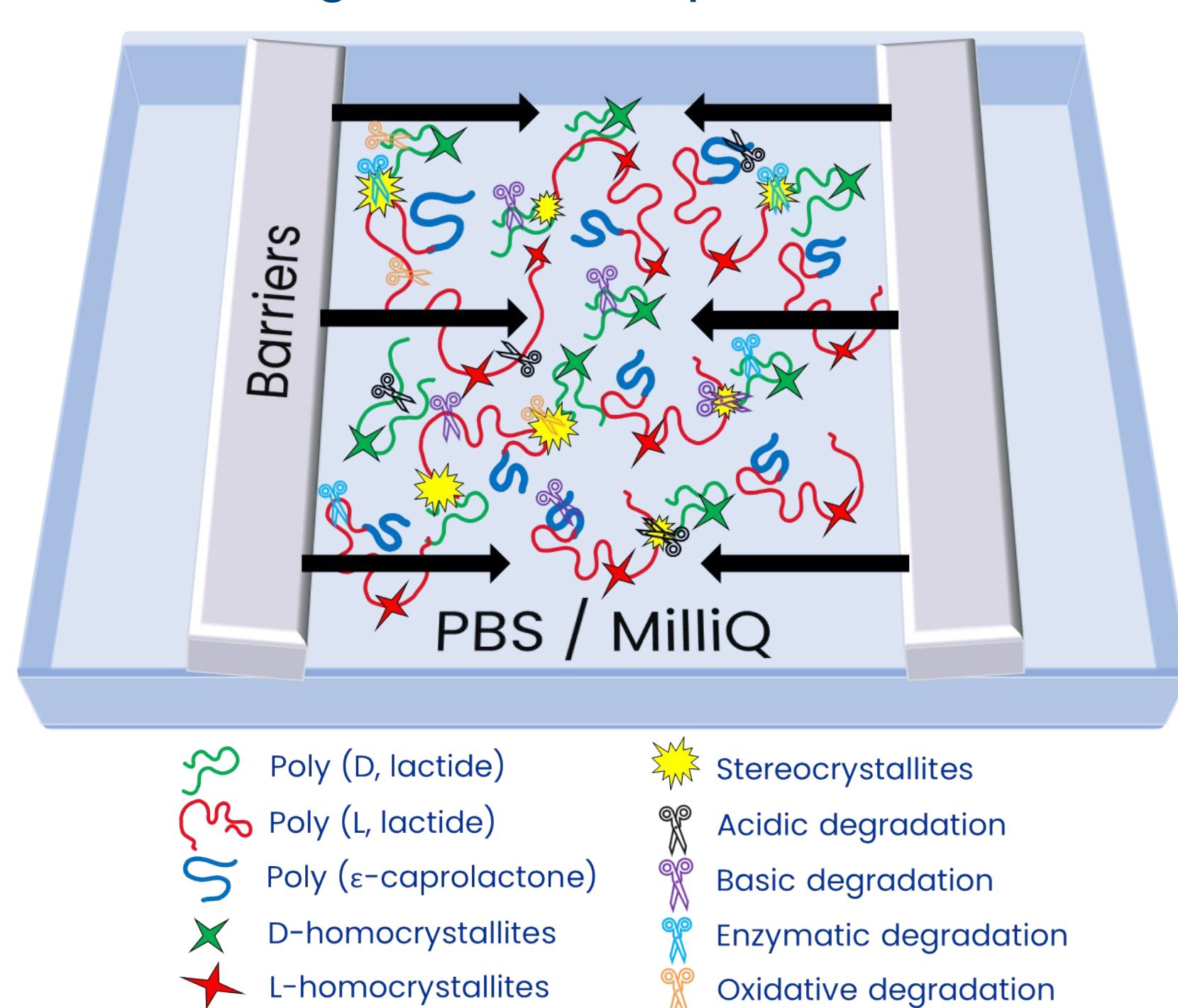


Figure 6: Schematic representation of the Langmuir degradation setup for PLLAcOCL, PDLA and blends

Formation of a monolayer of amphiphilic molecules over a liquid surface

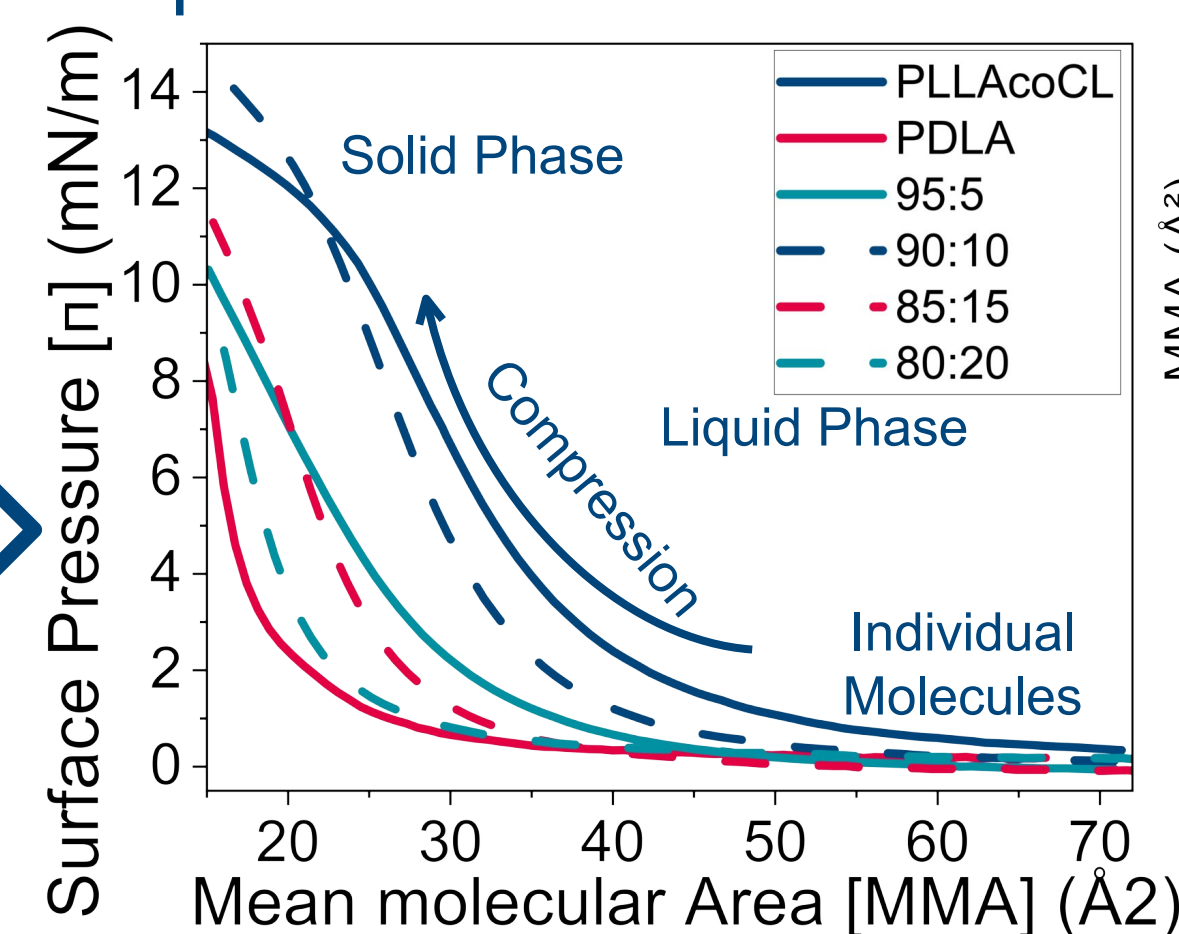


Figure 7: Increase in surface pressure upon compression, molecules change from individual molecules state to solid phase

➤ Degradation was observed under basic conditions but not in acidic, oxidative or enzymatic conditions

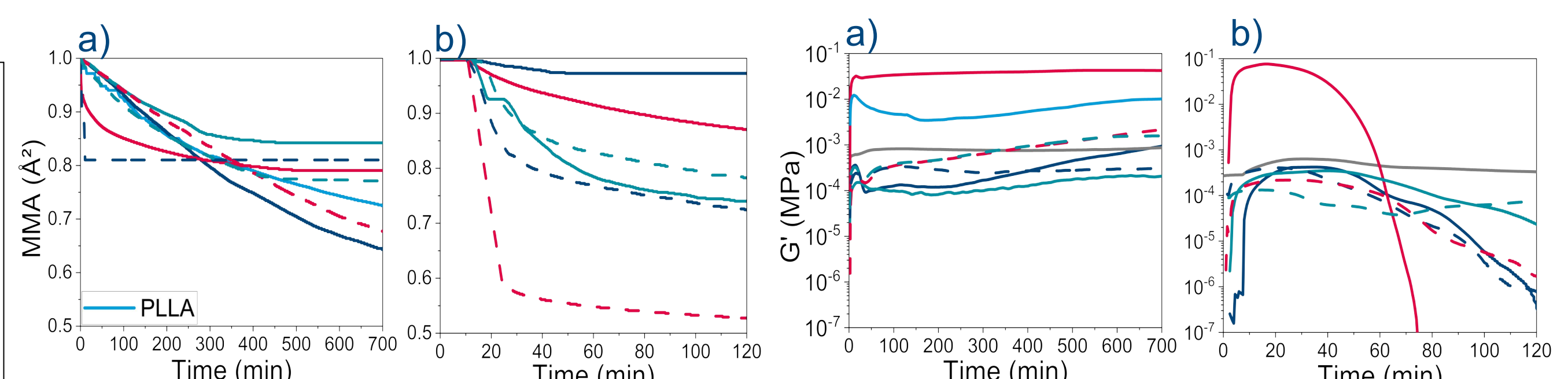


Figure 8: Surface pressure decreases over time in a) enzymatic and b) basic conditions

Degradation or formation of crystals upon compression

Confirmed by

Figure 9: Interfacial rheology results; an increase in G' correlates to crystallization of films in enzymatic conditions (a), decrease in G' correlates to degradation under basic conditions (b)

Conclusion and Outlook

- Sample composition (i.e. blending ratios) influences both degradation rate and mechanism, this can be tailored to the intended application
- The investigated degradation of the polymer system implies a promising potential of PLA-based materials being applied in cardiac implants and coverings
- Bulk degradation of different blends is in progress; allowing the ability to correlate thin film versus bulk degradation depending on the blend
- Data obtained will serve as input data for degradation material models to develop digital twins
- Applications as cardiac implants coverings will be further explored

References

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