

Supporting Information

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Hydrolytic degradation of PCL-PLLA semi-IPNs exhibiting rapid, tunable degradation

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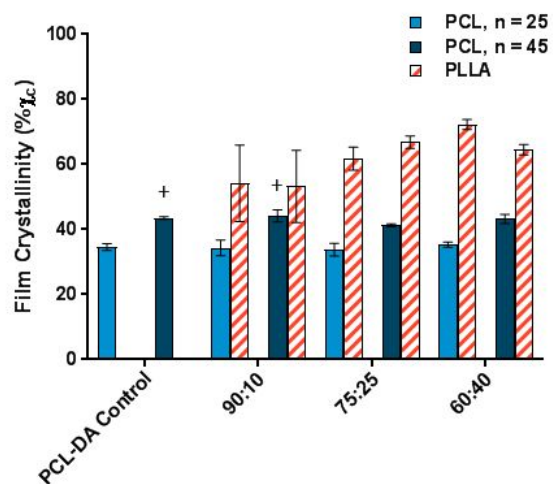


Fig. S1 PCL and PLLA weight-adjusted % crystallinity (% X_c) values for initial PCL-DA control and PCL-PLLA semi-IPN films as quantified from respective DSC curves (⁺ $p < 0.05$ vs corresponding PCL-DA ($n = 25$) film).

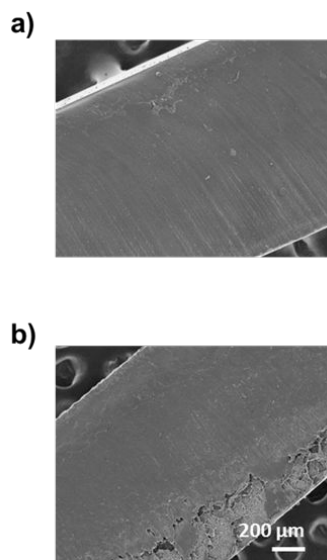


Fig. S2 SEM images of initial cross-sections for representative (a) PCL-DA ($n = 45$) control and (b) semi-IPN (75:25 [PCL:PLLA wt%; $n = 45$]) films.

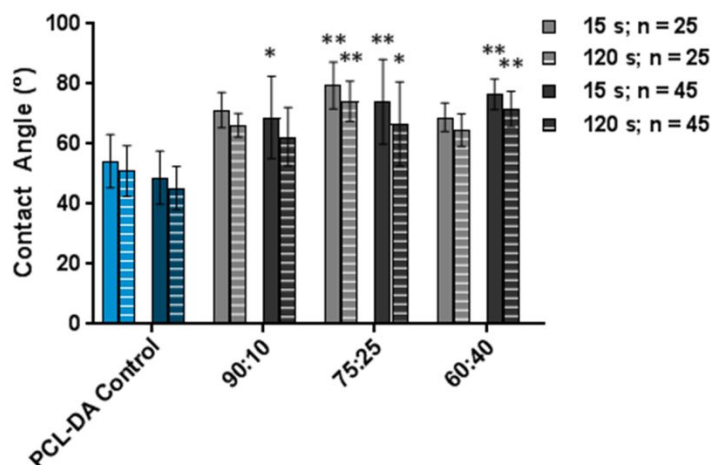


Fig. S3 Contact angle measurements for initial PCL-DA control and PCL-PLLA semi-IPN films indicating an increased hydrophobicity with PLLA (* $p < 0.05$, ** $p < 0.01$ vs corresponding PCL-DA control).

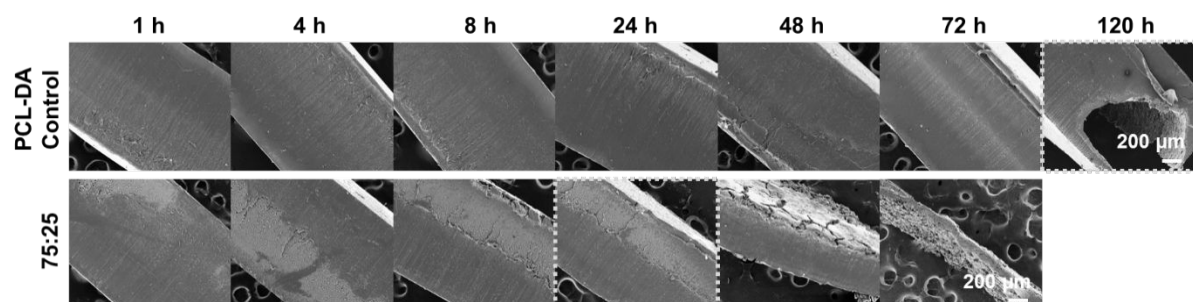


Fig. S4 SEM images of film cross-sections during accelerated (1 M NaOH, 37 °C) degradation for PCL-DA ($n = 25$) control and PCL-PLLA (75:25 [PCL:PLLA wt%]; $n = 25$) semi-IPN films. Films of similar mass loss (~41-54%) are boxed for comparison.

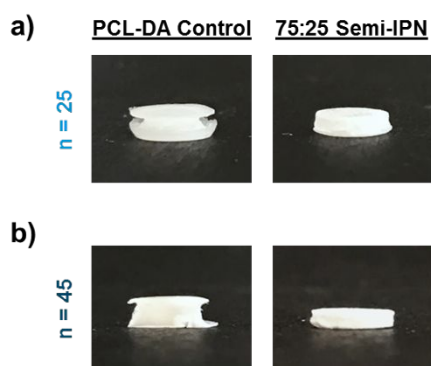


Fig. S5 Visual comparison of PCL-DA control and PCL-PLLA semi-IPN (75:25 [PCL:PLLA wt%]) cross-sections during accelerated (1 M NaOH, 37 °C) degradation, at similar mass loss (~41-54%; ~71-81%), for films based on (a) PCL-DA ($n = 25$) and (b) PCL-DA ($n = 45$).

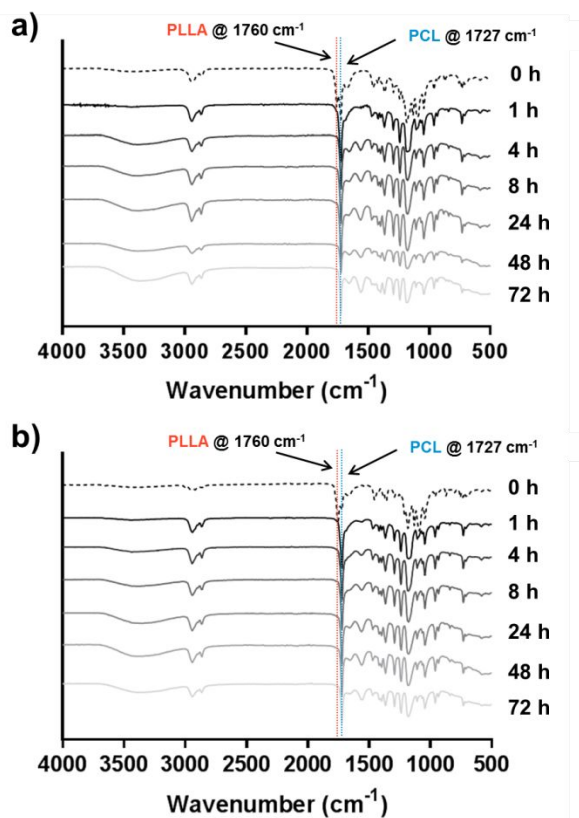


Fig. S6 ATR-FTIR spectra of PCL-PLLA (75:25 [PCL:PLLA wt%]) semi-IPN film surfaces upon accelerated (1 M NaOH, 37 °C) degradation when based on **(a)** PCL-DA ($n = 25$) and **(b)** PCL-DA ($n = 45$).

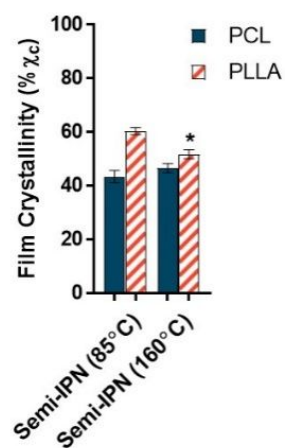


Fig. S7 Crystallinity comparison for representative 75:25 (PCL:PLLA, wt.%) semi-IPN (PCL-DA $n = 45$) PCL demonstrating reduced PLLA crystallinity in semi-IPN films annealed at 160°C (* $p < 0.05$).

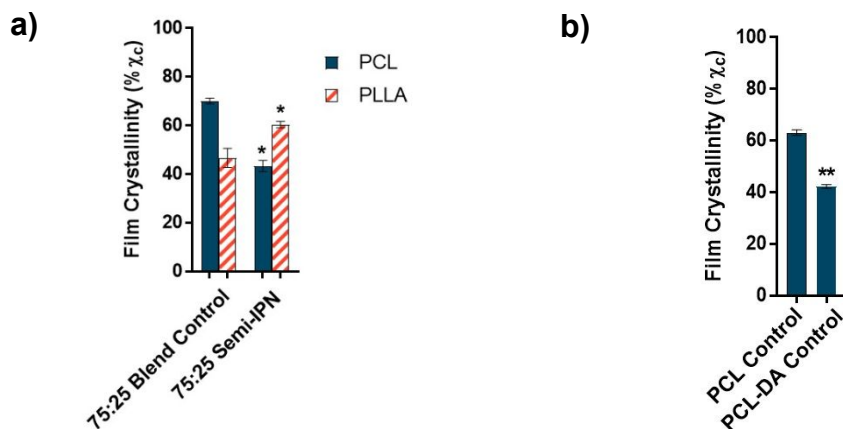


Fig. S8 Crystallinity comparison (PCL $n = 45$) for **(a)** representative 75:25 (PCL:PLLA, wt.%) showing significantly higher PCL crystallinity (* $p < 0.05$) and reduced PLLA crystallinity (* $p < 0.05$) in blends, and **(b)** showing significantly higher PCL crystallinity in PCL thermoplastic compared to cross-linked PCL-DA (** $p < 0.0001$)

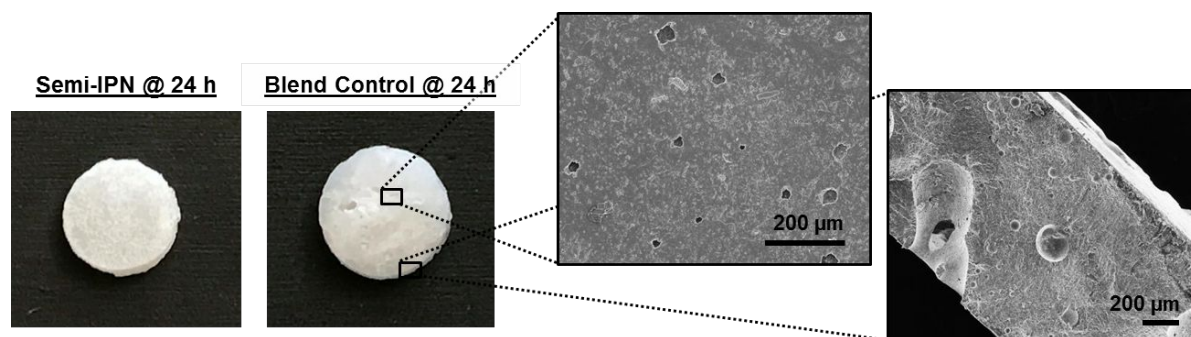


Fig. S9 Visual, close-up comparison of PCL-PLLA semi-IPN (75:25 [PCL:PLLA wt%]) and blend (75:25 [PCL:PLLA wt%]) control films at 24 h accelerated (1 M NaOH, 37 °C) degradation for representative films based on PCL-DA ($n = 25$).

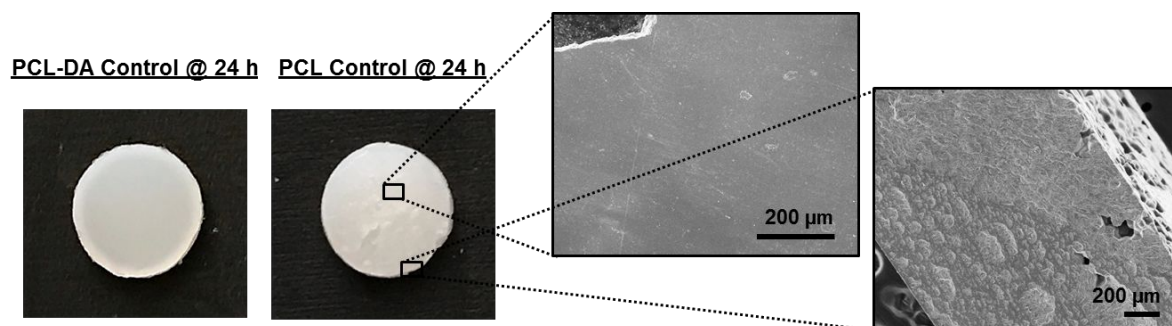


Fig. S10 Visual, close-up comparison of PCL-DA and PCL control films at 24 h accelerated (1 M NaOH, 37 °C) degradation for representative films based on PCL-DA ($n = 25$).

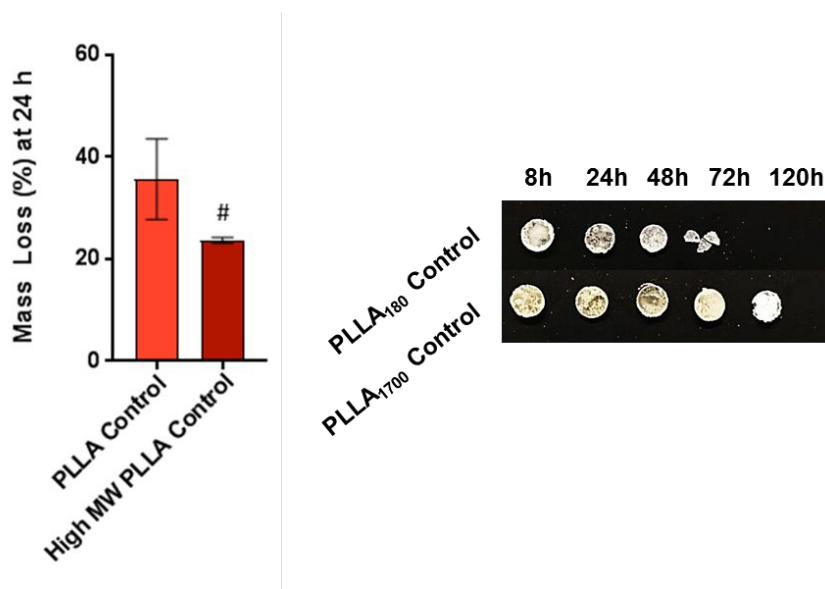


Fig. S11 The mass loss at 24 h under accelerated conditions (1 M NaOH, 37 °C) and visual degradation of PLLA control and high MW PLLA control films ([#] $p > 0.05$ vs PLLA control).

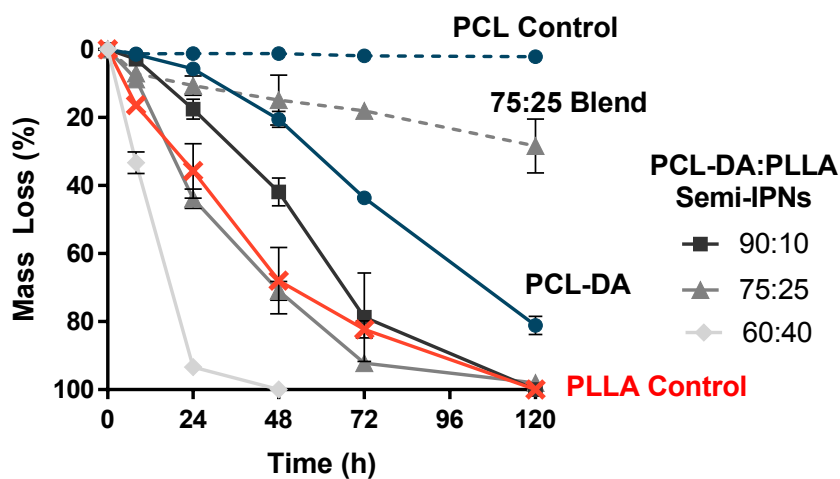


Fig. S12 The mass loss under accelerated conditions (1 M NaOH, 37 °C) for PCL control (n = 45), PLLA control (m = 90), PCL-DA (n = 45), corresponding blend and semi-IPNs (PCL-DA, n = 45).