Supporting Information

Interface-Modulated Morphological Transition of Biodegradable Poly(εcaprolactone) Crystals



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Figure S1. BAM images for $X_{PtBA} \sim 0.14$ PCL/PtBA blends at 22.5 °C obtained during isobaric area relaxation experiments. Experiments were performed at (Π /mN·m⁻¹): (A) 11, (B) 10.5; (C) 10.3, (D) 10, (E) 9.5, and (F) 8.5. BAM images were taken at <A> ~ 8-10 Å²·monomer⁻¹ and also represent different crystallization time (time/hour): (A) 0.3, (B) 0.7; (C) 1.5, (D) 2.7, (E) 6.8, and (F) 7.6. Solid-like domains appear bright in all of the 4.8 × 2.6 mm² BAM images.



Figure S2. BAM images for a PCL/PtBA blend of XPtBA ~ 0.14 obtained during an isobaric area relaxation experiment at $\Pi \sim 10.5 \text{ mN} \cdot \text{m}^{-1}$. BAM images were taken during isobaric experiments at (Time /hour): (A) 0.14, (B) 0.25, (C) 0.37, (D) 0.59, (E) 0.66 and (F) 0.70. Solid-like domains appear bright in all of the 4.8 × 2.6 mm² BAM images.



Figure S3. OM images of a single layer LS-film for a $X_{PtBA} \sim 0.14$ PCL/PtBA blend. The LS films were transferred onto PS coated silicon substrates at $\langle A \rangle \sim 5$ Å²·monomer⁻¹ during an isobaric area relaxation experiment at $\Pi \sim 11$ mN·m⁻¹: (A) Symmetric dendritic branching, a "winning" branch is indicated by the arrow; and (B) Detailed features of dendritic tips. A dendritic tip is indicated by the arrow.



Figure S4. OM images of a single layer LS-film for a $X_{PtBA} \sim 0.14$ PCL/PtBA blend. The LS films were transferred onto PS coated silicon substrates at $\langle A \rangle \sim 5$ Å²·monomer⁻¹ during an isobaric area relaxation experiment at $\Pi \sim 10.5$ mN·m⁻¹: (A) Symmetric dendritic branching, a "winning" branch is indicated by the arrow; and (B) Detailed features of a dendritic tip.



Figure S5. OM images of a single layer LS-film for a $X_{PtBA} \sim 0.14$ PCL/PtBA blend. The LS films were transferred onto PS coated silicon substrates at $\langle A \rangle \sim 5$ Å²·monomer⁻¹ during an isobaric area relaxation experiment at $\Pi \sim 10.3$ mN·m⁻¹: (A) Dendritic branches in one {100} sector are indicated by the arrow; and (B) Detailed features of a dendritic tip.



Figure S6. OM images of a single layer LS-film for a $X_{PtBA} \sim 0.14$ PCL/PtBA blend. The LS films were transferred onto PS coated silicon substrates at $\langle A \rangle \sim 5$ Å²·monomer⁻¹ during an isobaric area relaxation experiment at $\Pi \sim 10$ mN·m⁻¹: (A) Symmetric dendritic branching, dendritic branches in one {100} sector are indicated by the arrow; and (B) Detailed features of a dendritic tip.



Figure S7. OM images of a single layer LS-film for a $X_{PtBA} \sim 0.14$ PCL/PtBA blend. The LS films were transferred onto PS coated silicon substrates at $\langle A \rangle \sim 5$ Å²·monomer⁻¹ during an isobaric area relaxation experiment at $\Pi \sim 9.5 \cdot \text{m}^{-1}$. Both (A) and (B) show fat dendritic fingers.



Figure S8. OM image of a single layer LS-film for a $X_{PtBA} \sim 0.14$ PCL/PtBA blend. The LS film was transferred onto a PS coated silicon substrates at $\langle A \rangle \sim 8$ Å²·monomer⁻¹ during an isobaric area relaxation experiment at $\Pi \sim 8.5$ mN·m⁻¹.