Supplementary Information

Understanding nucleation efficiency of stereocomplex-crystallites on homochiral crystallization in poly (L-lactide)/poly (D-lactide) blends: Homogenization near crystal growth front

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1. Supplemental Methods

1.1. *In situ* small- and wide-angle X-ray scattering

In situ small- and wide-angle X-ray scattering (SWAXS) analysis was performed using a Nano-inXider (λ = 0.154 nm, Xenocs, Sassenage, France) operated at 50 kV, 0.6 mA, beam size 800 μm, and q range from 0.01 to 4 $Å^{-1}$. The solvent-casted films prepared by the method described in the main context were measured with an exposure time of 600 s. To study the melting and crystallization behavior of SCPLA during heating, the solvent-casted films were placed in a heating plate (HFSX350, Linkam Scientific Instruments Ltd., Surrey, United Kingdom) under a vacuum and heated from 30 °C to 270 °C at 2 °C/min.

1.2. Fourier-Transform Infrared Imaging

FTIR spectra were recorded using a PerkinElmer Spotlight 400 FTIR microscopy system (Massachusetts, USA) equipped with a 16-pixel MCT (mercury cadmium telluride) array detector with a 25 μm pixel

size. The sample thickness was maintained at 15–20 μm. Spectra were collected in the range of 4000–650 cm^{-1} with a resolution of 4 cm^{-1}, averaging 8 scans per measurement. The system was equipped with a Linkam THMS600 hot stage (Surrey, UK) for temperature-controlled experiments.

The in-situ FTIR imaging protocol was based on the DSC procedure described in Section 3 of the main text. Briefly, samples were rapidly heated from 30 °C to 270 °C at 100 °C/min, held at 270 °C for 3 minutes, and then cooled to 30 °C at 10 °C/min. The films were then subjected to self-nucleation temperatures (Ta) of 190 °C and 210 °C for 5 minutes to generate different melt structures. The protocol was further modified to study isothermal HC crystallization under different Ta conditions. After cooling from 190 °C or 210 °C, the films were held at 140 °C for 30 minutes to monitor HC crystallization.

The collected spectra were processed using OriginPro software for baseline correction and deconvolution with Gaussian fitting. This approach allowed the spatially resolved identification of SC and HC domains and provided detailed insight into how pre-existing SC crystallites influence subsequent HC formation.

2. Supplemental results

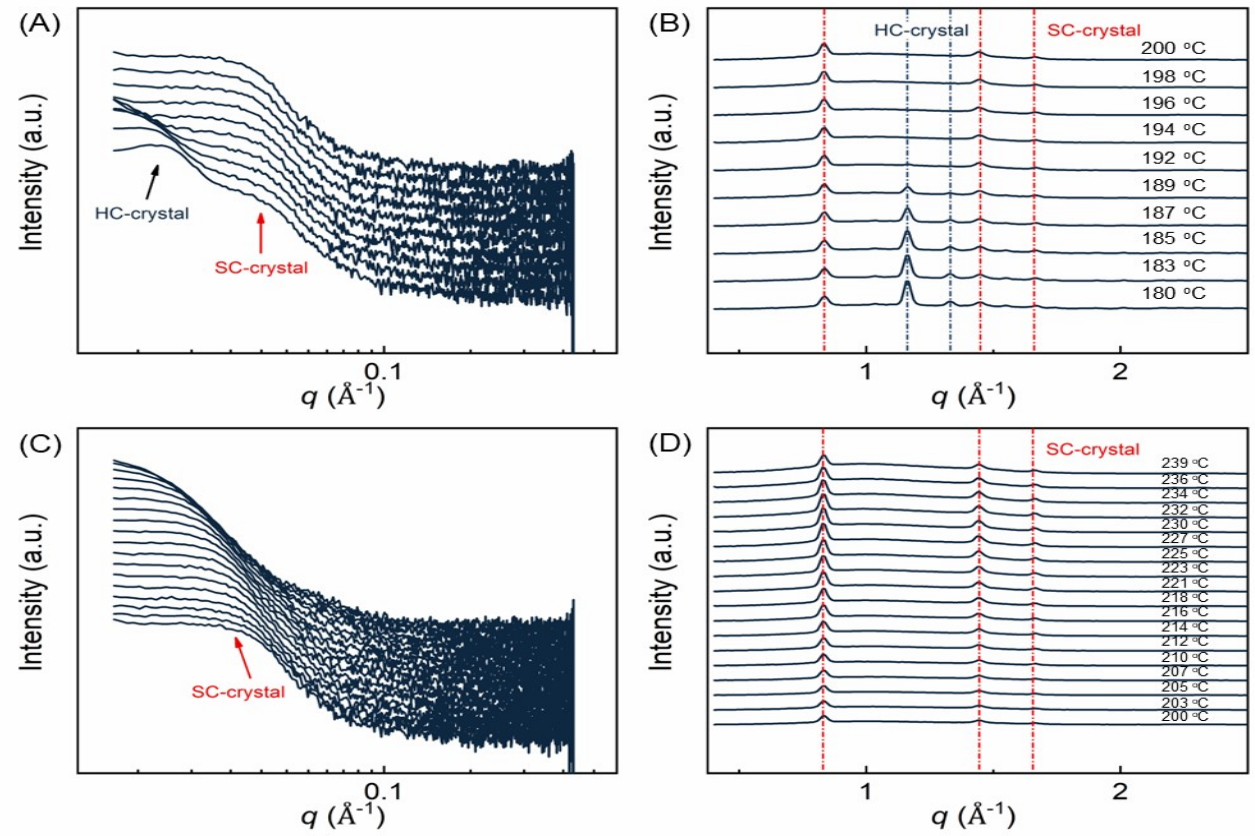
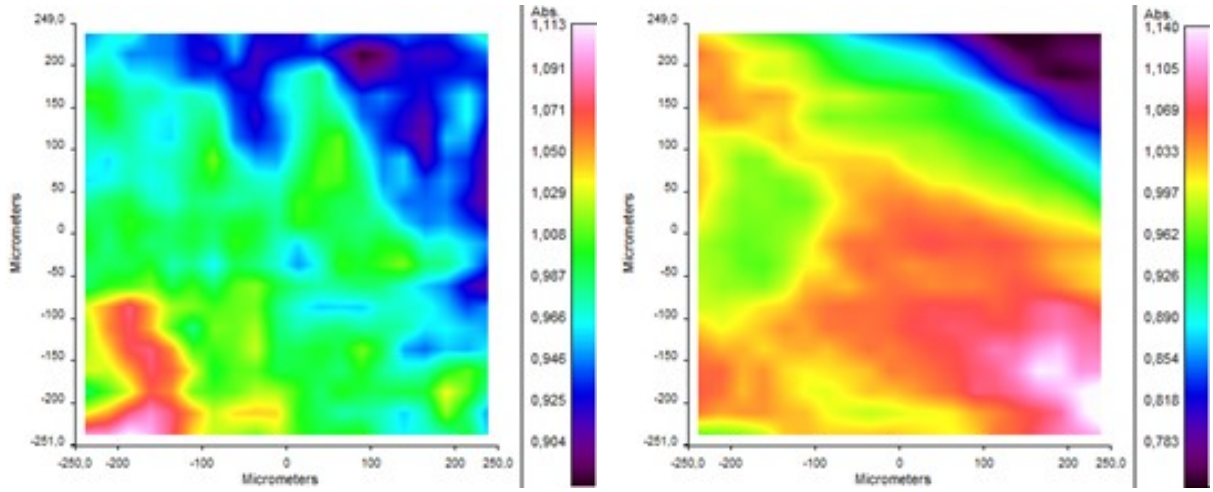


Figure 1S. (A) SAXS and (B) WAXS pattern of SCPLA during heating from 180 to 200 °C at 2 °C/min. The long spacing peaks and crystal diffraction of HC and SC-crystals in SAXS and WAXS patterns were marked correspondingly. (C) SAXS and (D) WAXS pattern of SCPLA during heating from 200 to 239 °C at 2 °C/min.



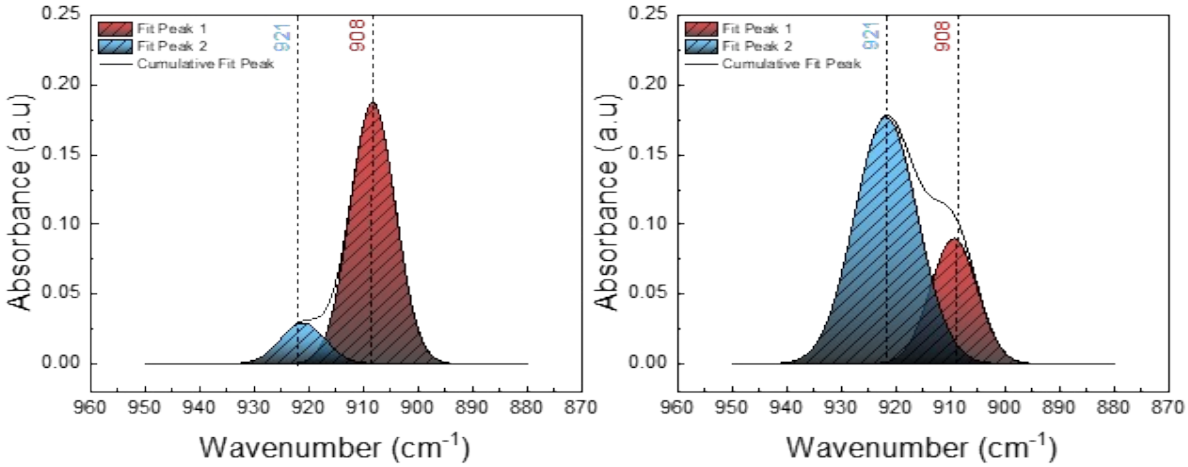


Figure 2S. FTIR images and their corresponding FTIR spectra of SCPLA crystallized at T_C 140 °C after 30 min following annealing at (A-C) T_a 220 °C, and (B-D) T_a 190 °C for 5 min.

Table 1S. Area calculations based on a Gaussian fit

		SCPLA crystallized at T_c 140 °C	
Samples	**SC peak area at 908**	**HC peak area at 921**	**SC peak area at 908**
SCPLA annealed at T_a 220 °C	3.13 ± 0.86	0.97 ± 0.10	2.55 ± 0.00
SCPLA annealed at T_a 190 °C	1.65 ± 0.35	1.94 ± 0.10	1.30 ± 0.10