

SUPPLEMENTARY INFORMATION

Pre-aggregation determines crystallinity in aliphatic polymer films deposited from supercritical fluids

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1. Grazing incidence wide angle x-ray scattering results

Figure S1 displays the azimuthal integration of the GIWAXS images displayed in Figure 3, showing scattering intensity as a function of scattering vector (q). The observed scattering pattern is consistent with the *i*PP α -phase,^{1, 2} with Miller indices of the reflections identified in red. In addition, a feature attributed to $\beta(300)$ is observed at 1.09 \AA^{-1} , indicating a small amount of the β -phase in *i*PP film grown at 3.5 MPa. This feature is not present at higher pressures.

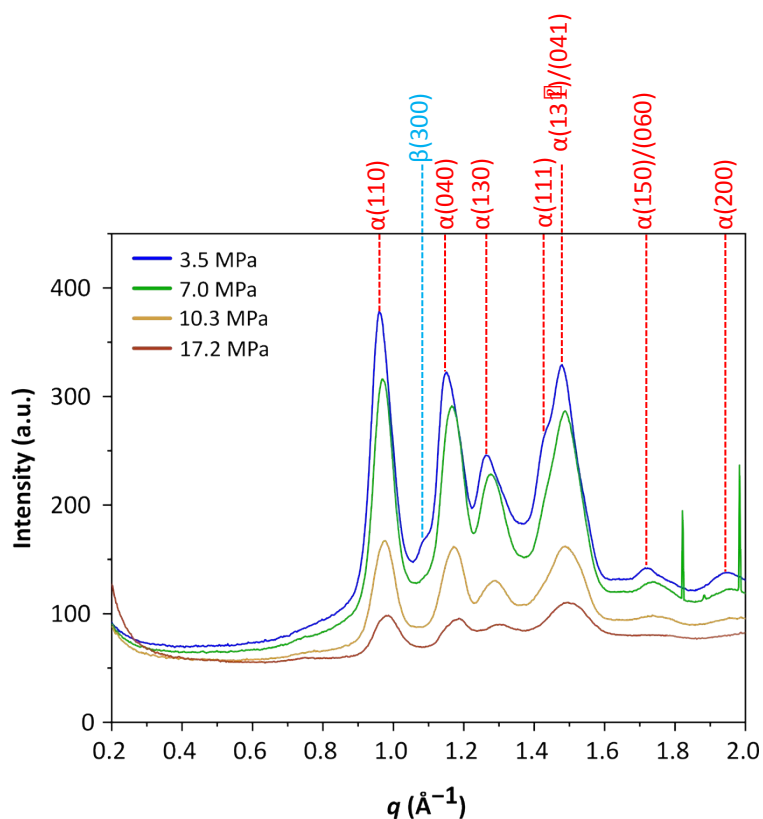


Figure S1. Azimuthally-integrated GIWAXS patterns for *i*PP films grown in *n*-pentane at different pressures.

Figure S2 shows the partial pole figures constructed from the GIWAXS patterns shown in Figure 3. The data show the relative orientation of the various lattice planes relative to the substrate normal ($\chi = 0^\circ$). The two most intense reflections are from the (110) and (040) lattice planes. The (110) reflection shows a mixture of in-plane and out-of-plane scattering, indicative of two major orientations; the (040) reflection is most intense in the out-of-plane direction (in the vicinity of $\chi \sim 0^\circ$). All reflections are consistent with one of the two major orientations (Figure 3), which both have the polymer chains oriented parallel to the surface.

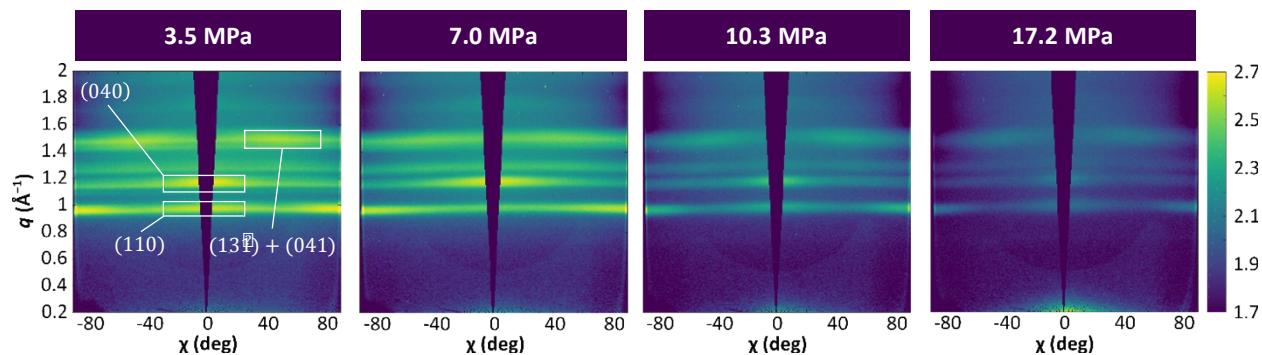


Figure S2. GIWAXS partial pole figures of the iPP films grown in n-pentane at different pressures.

2. References

1. A. T. Jones, J. M. Aizlewood and D. R. Beckett, *Die Makromolekulare Chemie*, 1964, **75 (1)**, 134-158.
2. G. Natta and P. Corradini, *Structure and Properties of Isotactic Polypropylene*, Pergamon, 1967.