

Electronic Supplementary Information

**Parallel micro- and mesophase transitions of thermoplastic
polyurethane induced by thermal annealing**

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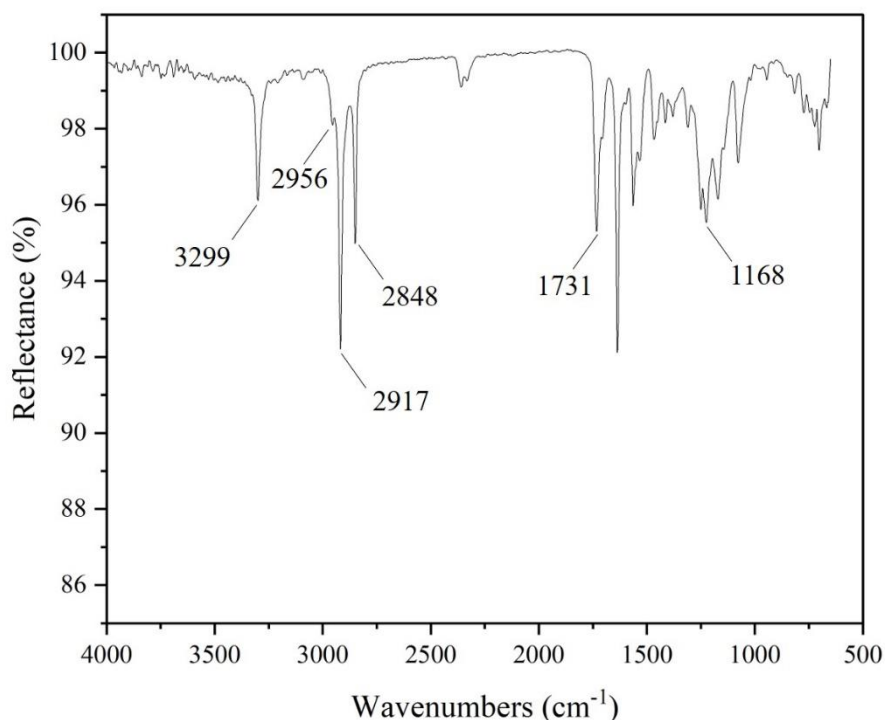


Fig. S1 FTIR spectra of as-received TPU (Elastollan® C85A10). The data were obtained at room temperature using Fourier transform infrared attenuated total reflection spectroscopy (FTIR-ATR) with a Nicolet iS50 FTIR spectrometer (Thermo Fisher Scientific Corp.) Prior to the measurement, the pristine granule sample was stored in a moisture-proof box at 25 °C. The spectra were collected over the range of 4000–550 cm^{-1} with 64 scans at a resolution of 2 cm^{-1} . The absorption peaks at 1731 cm^{-1} and 1168 cm^{-1} denote the stretching vibration of the C=O unit and the asymmetric stretching vibration of the C-O unit in the ester components of TPU, respectively. The absorption peak at 3299 is attributed to the N-H stretching vibrations.¹ For the soft segment polyether polyols, the three peaks near 2956 cm^{-1} , 2917 cm^{-1} , and 2848 cm^{-1} are assigned to the asymmetric stretching vibrations of C-H in methyl, in methylene and the symmetric stretching vibrations in methylene,² respectively.

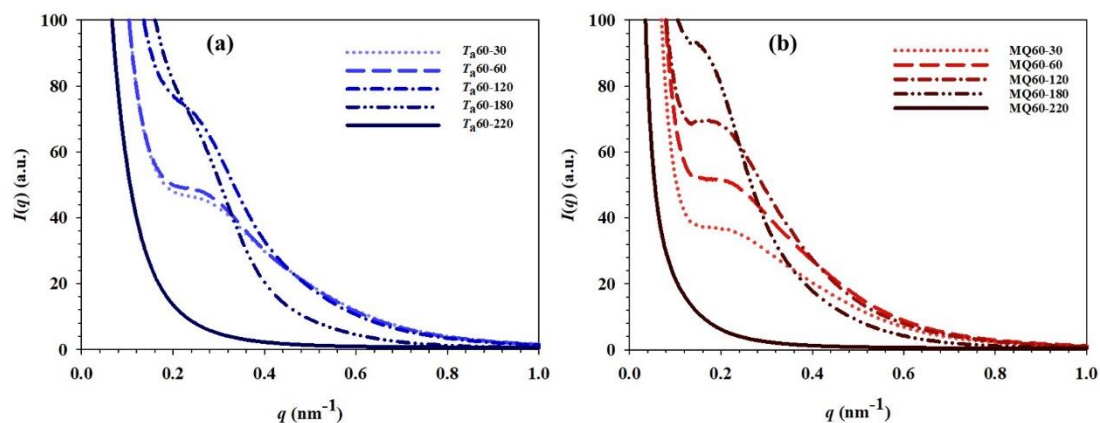
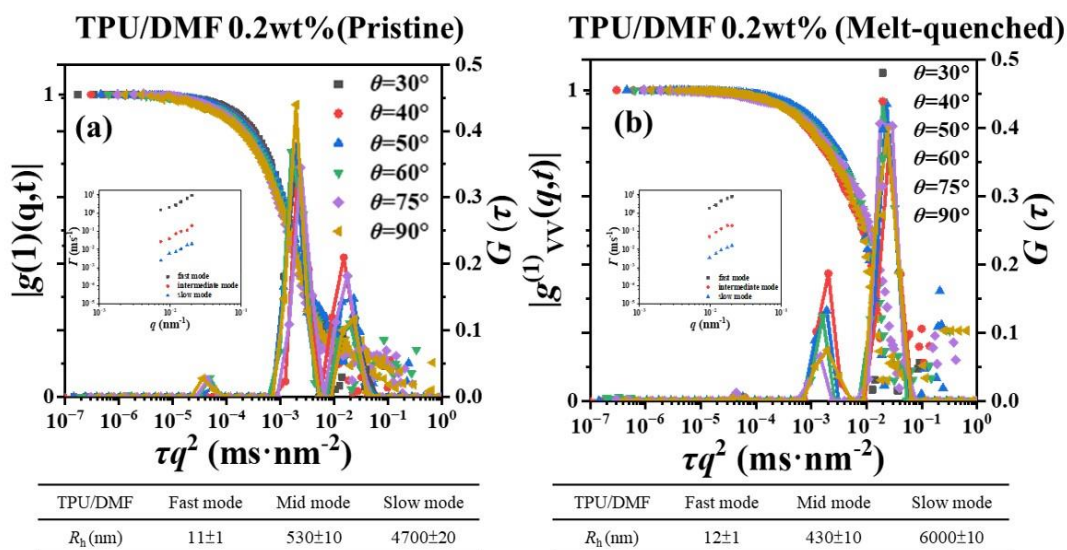


Fig. S2 Comparison of SAXS intensity profiles during the heating process for (a) pristine and (b) melt-quenched (melting at 220 °C for 15 min and then quickly quenched at -5 °C for 24 h) TPU samples subject to thermal annealing at 60 °C for 24 h, after which each sample was quickly cooled to 30 °C for the subsequent heating and measurement.



Fig. S3 Photo of the purified Elastollan® C85A10 sample.



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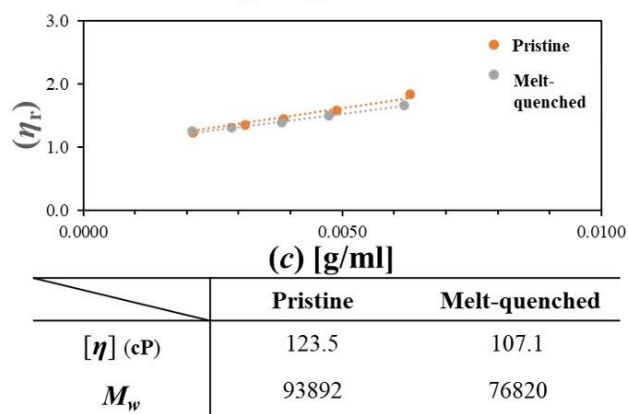


Fig. S4 DLS results on the 0.2 wt% solutions produced from (a) pristine TPU and (b) melt-quenched TPU dissolved in DMF. (c) Corresponding relative viscosities and polymer molecular weights M_w determined from the Mark-Houwink equation:³ $[\eta] = KM_w^\alpha$.

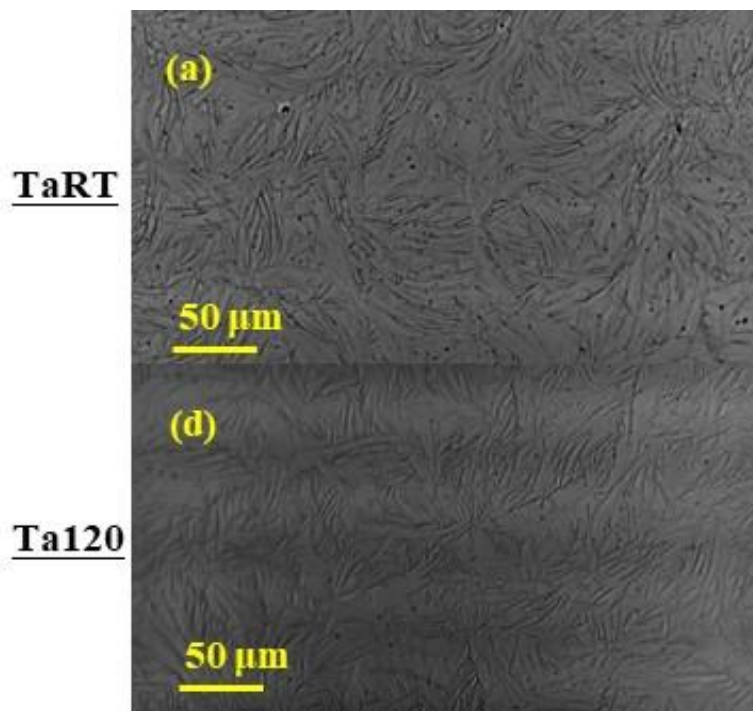


Fig. S5 OM images of the Pellethane 2363-80A sample shown in Fig. 7 of the main text after contrast enhancement.

References

- 1 A. Puentes-Parodi, M. Gedan-Smolka, A. Leuteritz, M. Gehde and I. Kuehnert, *J. Compos. Mater.*, 2018, **52**, 3669-3680.
- 2 L. Jiao, H. Xiao, Q. Wang and J. Sun, *Polym. Degrad. Stab.*, 2013, **98**, 2687-2696.
- 3 H. C. Beachell and J. C. Peterson, *J. Polym. Sci., Part A: Polym. Chem.*, 1969, **7**, 2021-2029.