<u>**Supporting Information**</u>

Toughened Isotactic Polypropylene: Phase Behavior and Mechanical Properties of Blends with Strategically Designed Random Copolymer Modifiers

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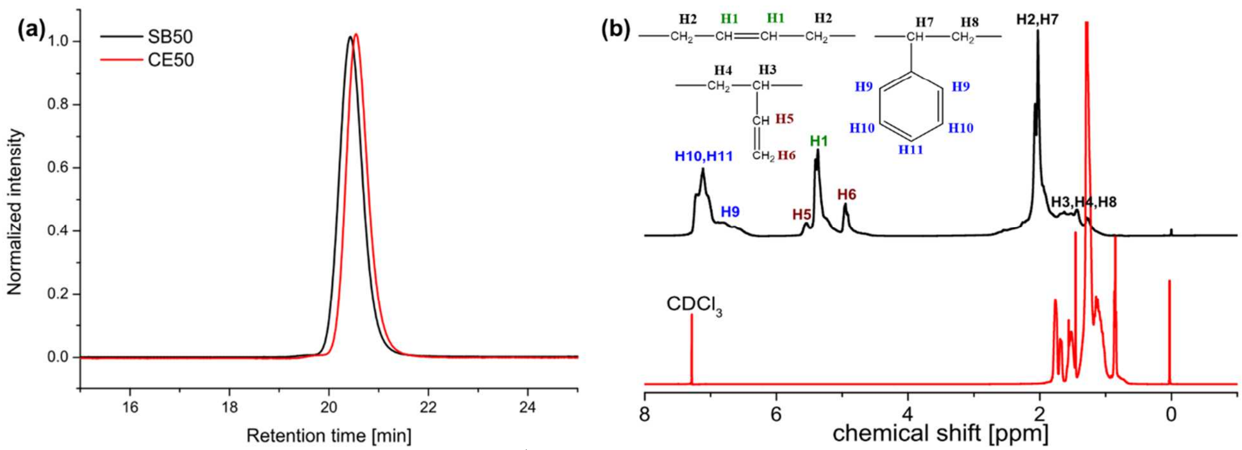

Figure S1. Representative SEC (a) and ^{1}H NMR traces (b) traces of CE50 (red) and unsaturated counterpart SB50 (black). The disappearance of peaks associated with aromatic hydrogen in the NMR spectrum and an invariant SEC trace indicate complete hydrogenation without degradation.

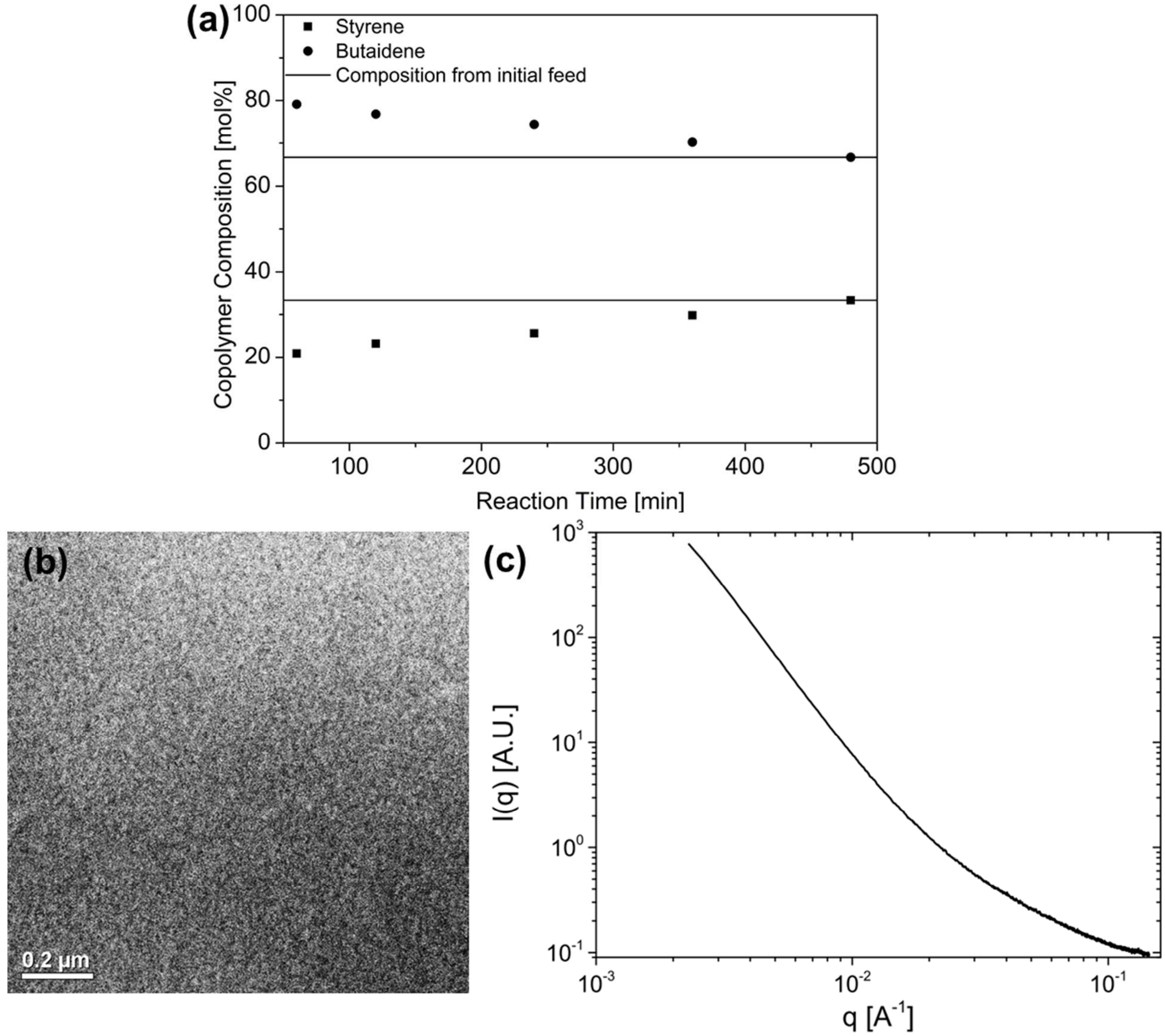

Figure S2. (a) SB50 copolymer composition as a function of reaction time taken at 1h, 2h, 4h, 6h and 8h of the reaction, (b) representative TEM image and (c) SAXS profile of SB50.

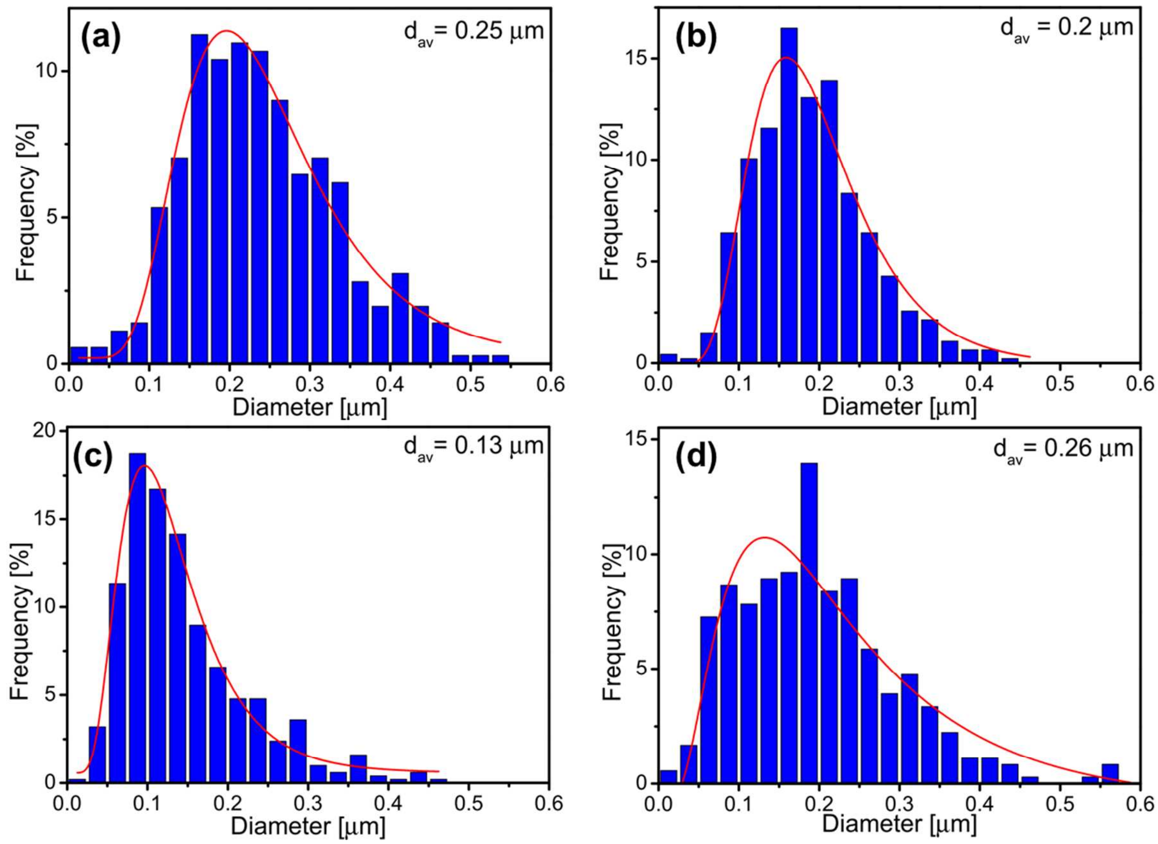

Figure S3. Histograms determined from SEM images of 5 wt.% loadings of (a) CE50, (b) CE60, (c) CE80 and (d) PE in *i*PP. The histograms were fitted with log-normal distribution (red solid line) and the average diameter (d_{av}) are provided in the histograms.

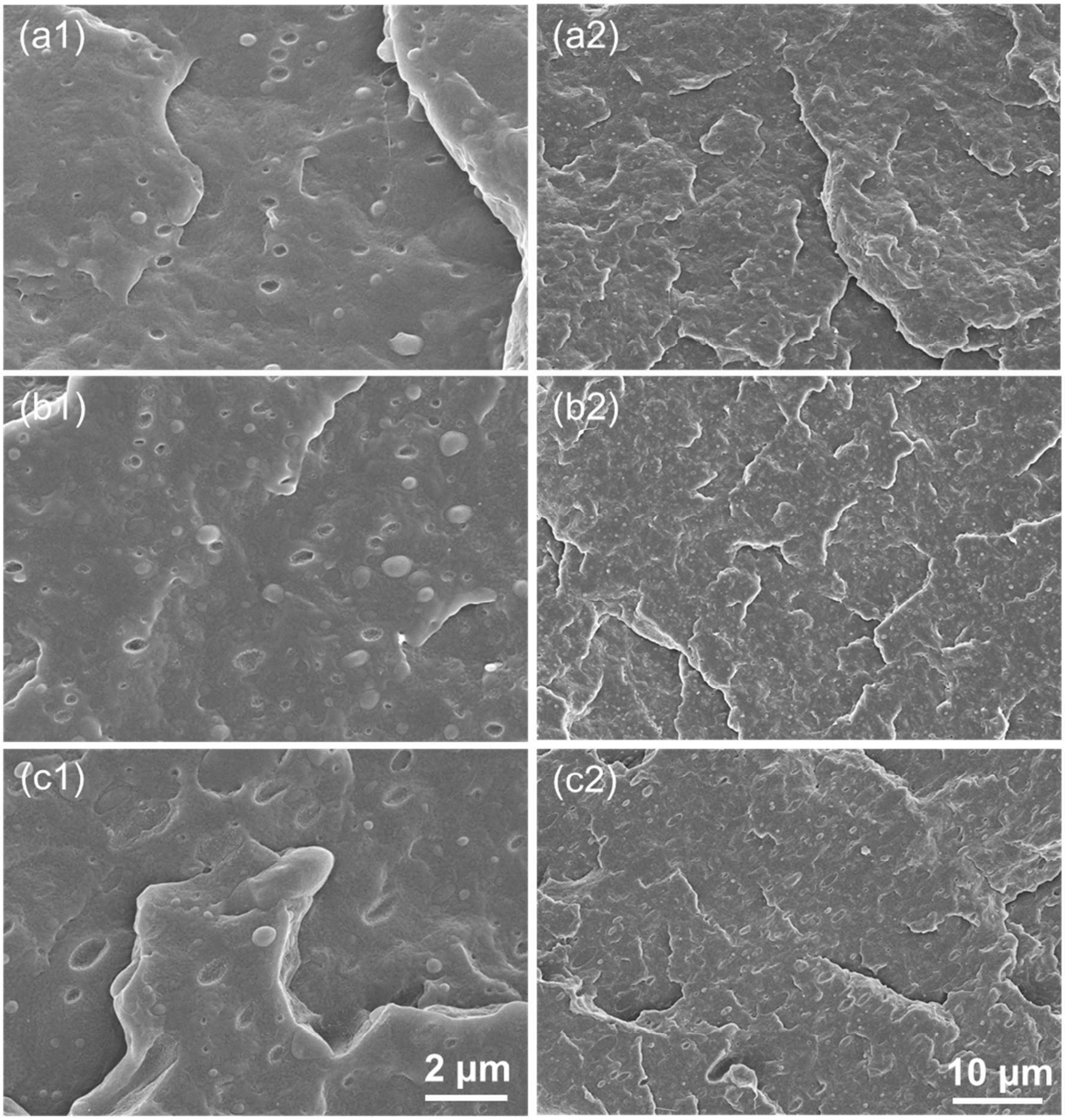

Figure S4. Representative SEM images of *i*PP blends containing (a1, a2) 5 wt.%, (b1, b2) 10 wt.%, (c1, c2) 20 wt.% CE50 at higher (1) and lower (2) magnification.

Table S1. Droplet size of *i*PP/CE copolymers blends at various loadings

Blends	CE copolymer weight percentage [%]	d_n [µm]	d_v [µm]	d_v/ d_n
***i*PP/PE**	5	0.19	0.34	1.8
	10	0.34	0.69	2.0
	20	0.83	1.9	2.3
***i*PP/CE50**	5	0.24	0.33	1.4
	10	0.28	0.46	1.6
	20	0.55	1.1	2
***i*PP/CE60**	5	0.19	0.26	1.4
	10	0.3	0.5	1.7
	20	0.56	1.0	1.9
***i*PP/CE70**	5	0.18	0.25	1.4
	10	0.2	0.36	1.8
	20	0.44	1.0	2.3
***i*PP/CE80**	5	0.15	0.27	1.8
	10	0.16	0.29	1.8
	20	0.52	1.2	2.3
***i*PP/PCHE**	5	2.5	8.2	3.3
	10	4.2	18	3.9
	20	4.6	20	4.4

Tables S2. Mechanical properties of the *i*PP/CE copolymers blends

Blends	CE copolymer weight percentage [%]	Elastic modulus *E* [GPa]	Yield stress σ_y* [Mpa]	Elongation at break ε_b [%]
***i*PP**	0	1.68 ± 0.07	30 ± 1	14 ± 5
***i*PP/PE**	5	1.60 ± 0.04	31 ± 1	93 ± 25
	10	1.52 ± 0.03	29 ± 1	26 ± 9
	20	1.32 ± 0.10	25 ± 1	20 ± 7
***i*PP/CE50**	5	1.64 ± 0.04	27 ± 1	346 ± 60
	10	1.54 ± 0.04	25 ± 1	432 ± 51
	20	1.32 ± 0.06	20 ± 2	480 ± 47
***i*PP/CE60**	5	1.62 ± 0.05	27 ± 1	357 ± 45
	10	1.53± 0.02	26 ± 1	430 ± 49
	20	1.40 ± 0.04	22 ± 1	509 ± 47
***i*PP/CE70**	5	1.67 ± 0.06	29 ± 2	415 ± 34
	10	1.72 ± 0.12	28 ± 1	375 ± 41
	20	1.68 ± 0.07	27 ± 1	390 ± 25
***i*PP/CE80**	5	1.76 ± 0.04	31 ± 1	19 ± 5
	10	1.82 ± 0.07	30 ± 2	25 ± 7
	20	1.77 ± 0.05	-	3 ± 0.4
***i*PP/PCHE**	5	1.85 ± 0.04	28 ± 1	19 ± 5
	10	1.79 ± 0.04	20 ± 1	10 ± 2
	20	1.82 ± 0.07	-	2 ± 0.3

* Unspecified yield stress indicates that the specimen broke before yielding.

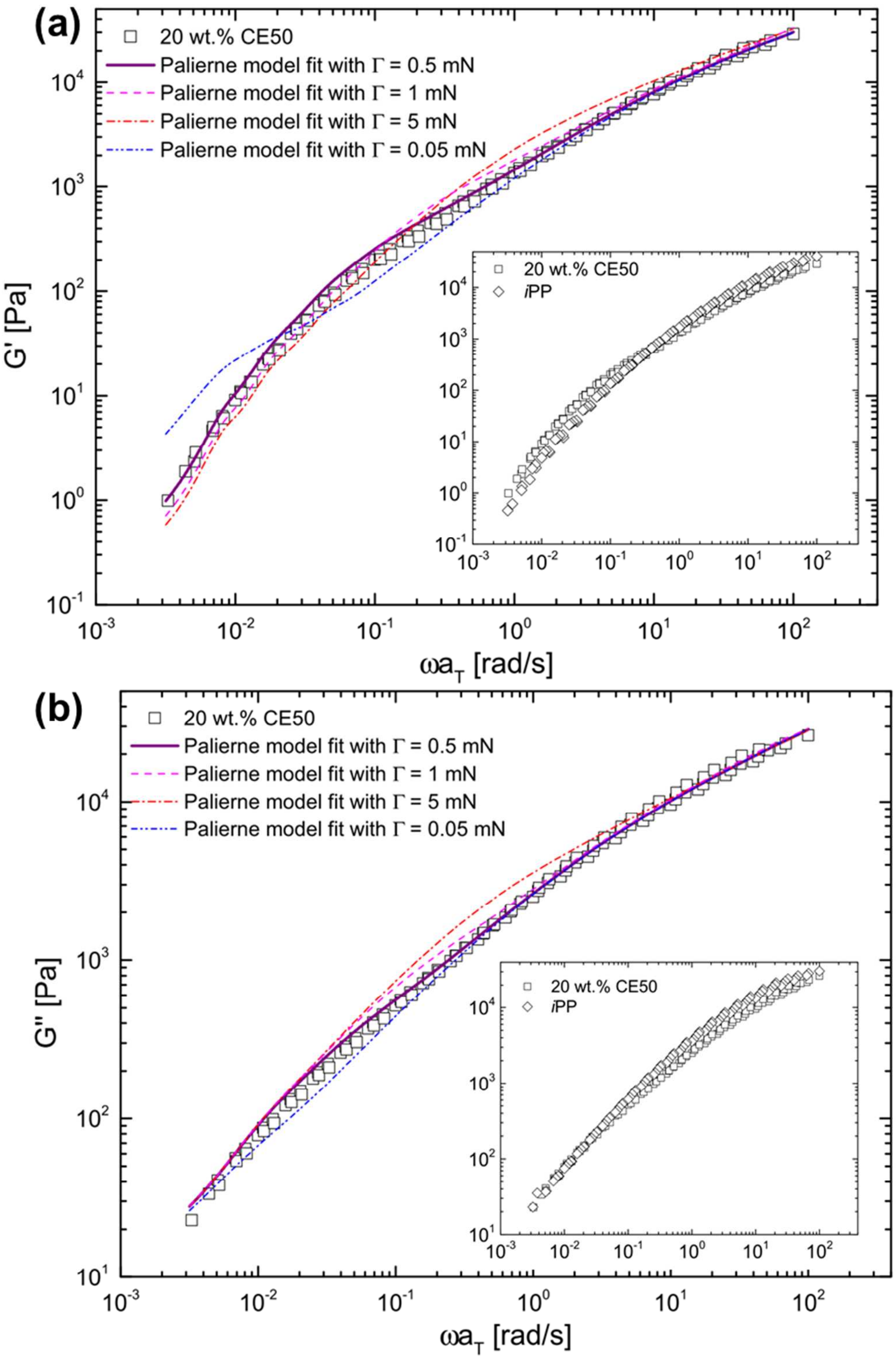

Figure S5. Comparison of the Palierne model predictions with experimental data for 20 wt.% CE50/*i*PP blends at 180 °C using different interfacial tensions; (a) storage modulus, (b) loss modulus. The insets show the storage and loss moduli of 20 wt.% CE50/*i*PP blends and *i*PP.

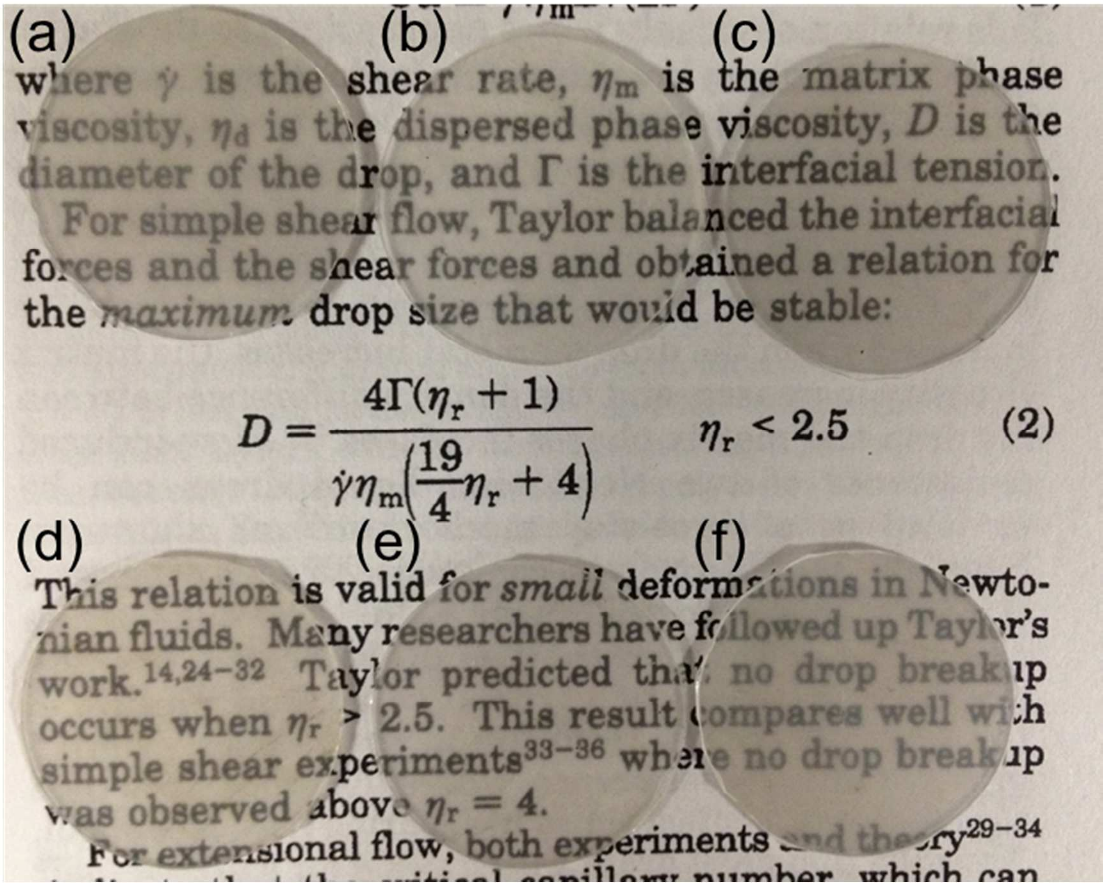

Figure S6 Transparency of 0.9 mm thick discs of (a) pure *i*PP, and 20 wt.% CE copolymers/*i*PP blends of (b) CE50, (c) CE60, (d) CE70, (e) CE80, and (f) PCHE.

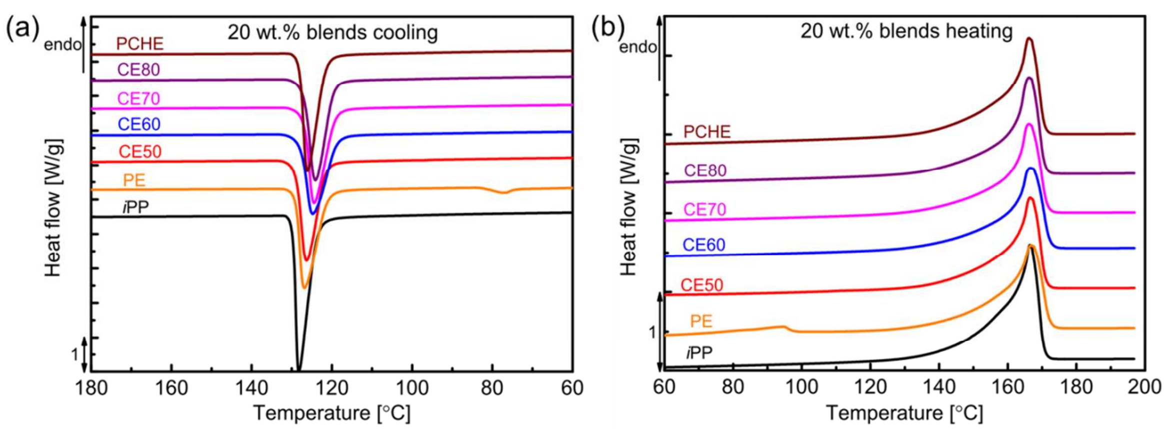

Figure S7. DSC traces of 20wt.% *i*PP/CE copolymers blends and pure *i*PP obtained while (a) cooling from 200 °C at a rate of 10 °C/min, and (b) heating at 10 °C/min following the cooling cycle. Curves were shifted vertically for clarity.

Table S3. Crystallization behavior of *i*PP and *i*PP/CE blends

Blends	CE copolymer weight percentage [%]	The onset crystallization temperature T_{on}[a] [°C]	The peak crystallization temperature T_{peak} [°C]	Crystallinity X_c [%]
***i*PP**	0	129.6	128.8	52
***i*PP/PE**	5	129.2	127.4	53
	10	129.1	127.1	53
	20	129.0	126.8	53
***i*PP/CE50**	5	128.3	126.9	48
	10	128.8	126.9	52
	20	129.0	126.4	51
***i*PP/CE60**	5	128.4	126.6	49
	10	128.1	126.6	51
	20	127.8	124.7	50
***i*PP/CE70**	5	128.1	126.4	49
	10	127.3	125.1	52
	20	126.9	124.4	51
***i*PP/CE80**	5	127.7	126.0	46
	10	127.4	124.8	55
	20	127.0	124.2	52
***i*PP/PCHE**	5	129.0	127.2	51
	10	128.8	127.1	50
	20	128.0	126.1	53

a. T_{on} is defined as the temperature as the intercept of the tangents at the baseline and the high temperature side of the exotherm.

b. Crystallinity is calculated using $X_c = \frac{\Delta H_{melt}}{\omega \cdot \Delta H_{fusion}}$ where ΔH_{melt} is the measured enthalpy of melting, ω is the weight fraction of *i*PP in the blend and ΔH_{fusion} = 207 J/g is the heat of fusion for pure crystalline *i*PP.

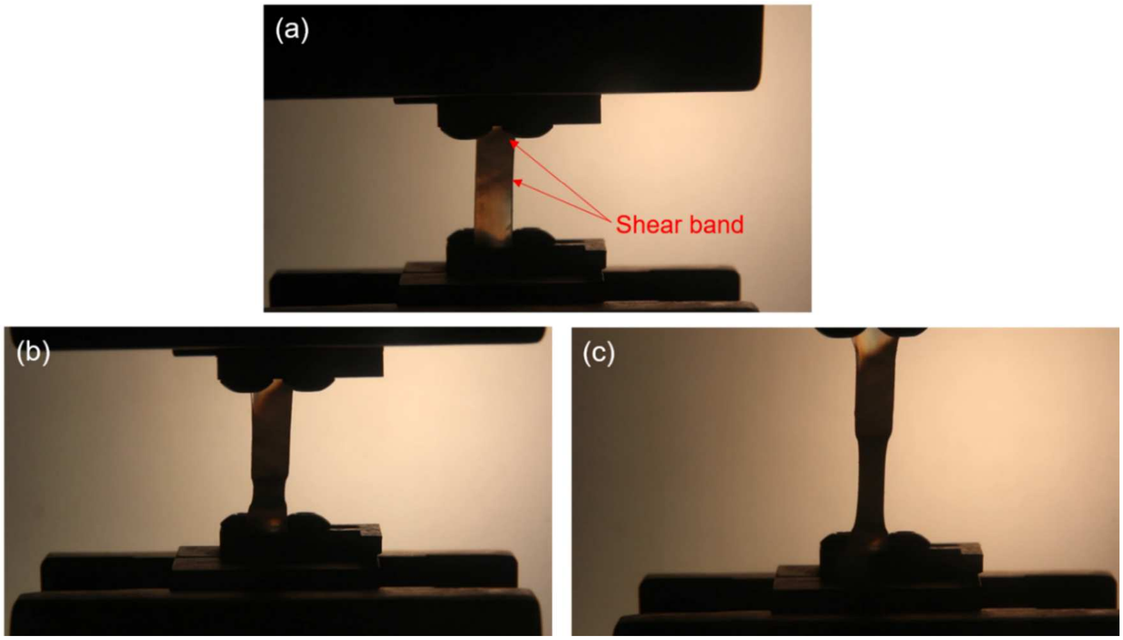

Figure S8. Representative images of three dumbell shape specimens containing 5 wt.% CE50 taken between polarizers at (a) the yield point, (b) 20% strain, and (c) 50% strain. The red arrows denote the formation of shear bands right after the yield point.

Table S4. The interparticle distance *l* of CE copolymer/*i*PP blends with different compositions

Blends	CE copolymer volume fraction φ	Average diameter d_n [μm]	Interparticle distance *l* [μm]
***i*PP/PE**	5.0	0.19	0.23
	10	0.34	0.25
	20	0.83	0.31
***i*PP/CE50**	5.0	0.24	0.29
	9.9	0.28	0.21
	20	0.55	0.20
***i*PP/CE60**	4.9	0.19	0.23
	9.8	0.30	0.22
	20	0.56	0.22
***i*PP/CE70**	4.9	0.18	0.22
	9.7	0.20	0.15
	20	0.44	0.17
***i*PP/CE80**	4.8	0.15	0.17
	9.6	0.16	0.12
	19	0.52	0.20
***i*PP/PCHE**	4.8	2.5	3.1
	9.5	4.2	3.2
	19	4.6	1.7