Supporting Information for

Pressure-Sensitive Adhesives from Renewable Triblock Copolymers

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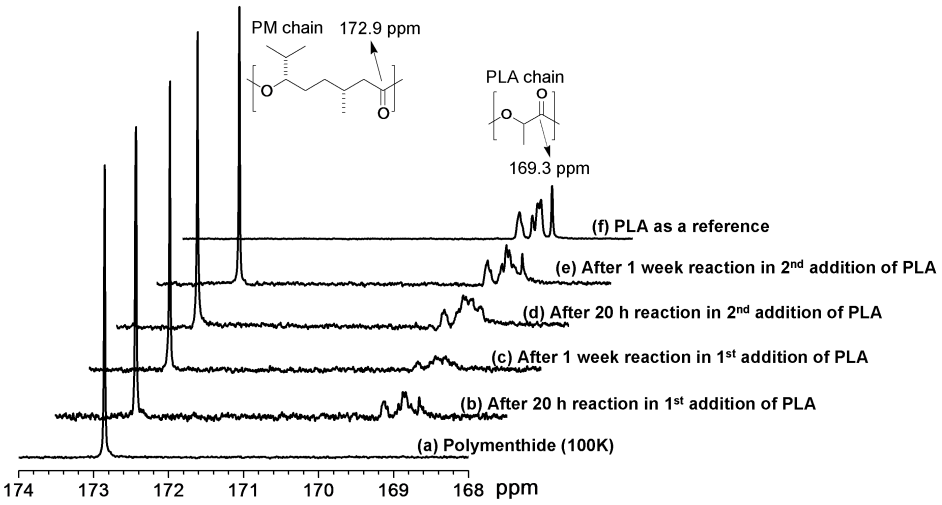

Figure S1. ^{13}C NMR spectra showing the carbonyl carbons of (a) PM(100)B as a macroinitiator; (b) PLA-PM-PLA(10-100-10)B by one-pot, two step process; (c) PLA-PM-PLA(10-100-10)B after one week at 135 °C; (d) PLA-PM-PLA(20-100-20) prepared by one more addition of lactide via the one-pot, two step process; (e) PLA-PM-PLA(20-100-20) after one week at 135 °C; and (f) PLA (as a reference; poly(D,L-lactide)).

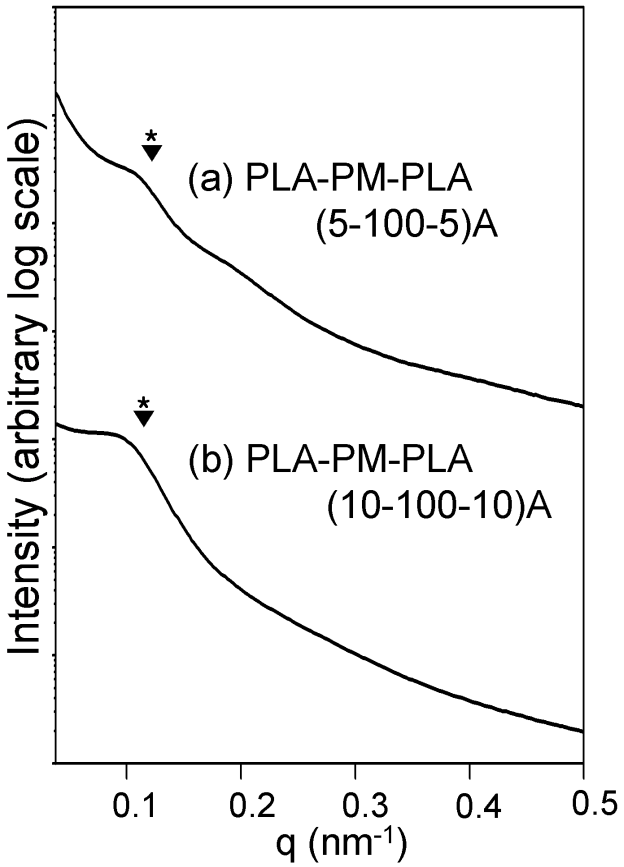

Figure S2. One-dimensional SAXS profiles for PLA-PM-PLA triblock copolymers at 25 °C.

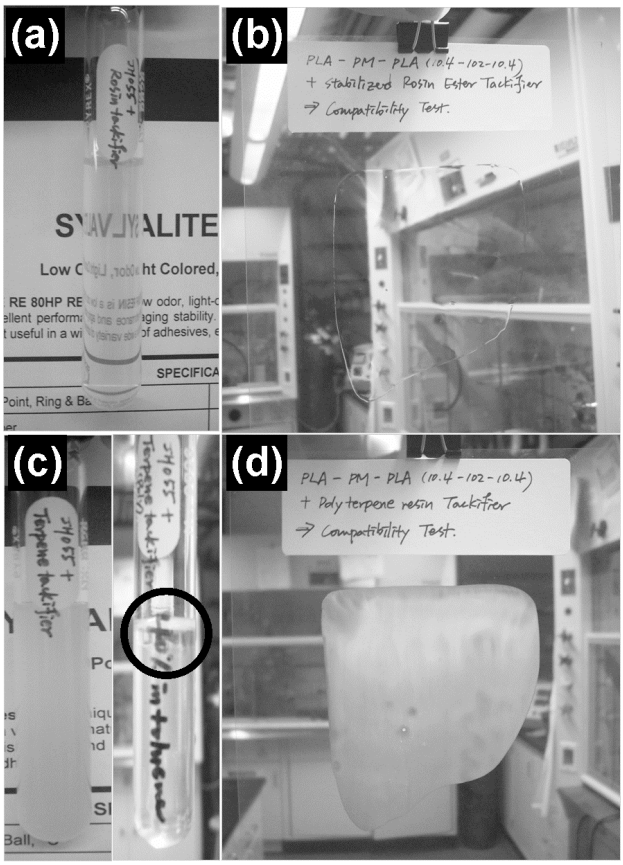

Figure S3. Compatibility tests. A mixture of PLA-PM-PLA(10-100-10)A/rosin ester (PE) tackifier in toluene (a) a transparent film from the PLA-PM-PLA(10-100-10)A/RE tackifier (b) a mixture of PLA-PM-PLA(10-100-10)A/polyterpene (PT) tackifier in toluene (c) a non-transparent film from the PLA-PM-PLA(10-100-10)A/PT tackifier solution (d). The tubes in (a) and (c) were allowed to stand for 10 days after mixing.

Compatibility Tests of Renewable Triblock copolymers/Tackifier Systems. Experiments were conducted to probe the compatibility of the renewable PSA systems using solution and film methods (Figure S2). Solution tests were carried out on the mixtures of PLA-PM-PLA(10-100-10)A and tackifier (rosin ester (RE) or polyterpene (PT)) in toluene. The test tubes were allowed to stand for 10 days after mixing for 3 h. The triblock copolymer and RE

tackifier solution was a transparent and homogeneous solution even after 10 days (a), while the blend of the triblock copolymer and PT tackifier was opaque (c). After 10 days, the mixture separated into two phases (right figure of (c) having black circle) with the upper and lower portions corresponding to polyterpene-rich and elastomer-rich phases. The transparency of a film from the triblock copolymer and both tackifiers on PET sheets was also tested. The film of the triblock copolymer and RE tackifier blend showed clear transparency (b), while the film was turbid for the triblock copolymer/PT mixture (d). In addition, PLA-PM-PLA and RE were compatible on the film surface even in the absence of toluene. These results indicate that PLA-PM-PLA/RE system showed excellent compatibility, which was superior to the PLA-PM-PLA/PT system. Therefore, we decided to choose the PLA-PM-PLA/RE system for thermoplastic properties and adhesive evaluations. Eight PSA samples were prepared by the combination of two triblock copolymers, PLA-PM-PLA(5-100-5 and 10-100-10)A with 0, 20, 40, and 60 wt % of RE based on solvent-based process using toluene.

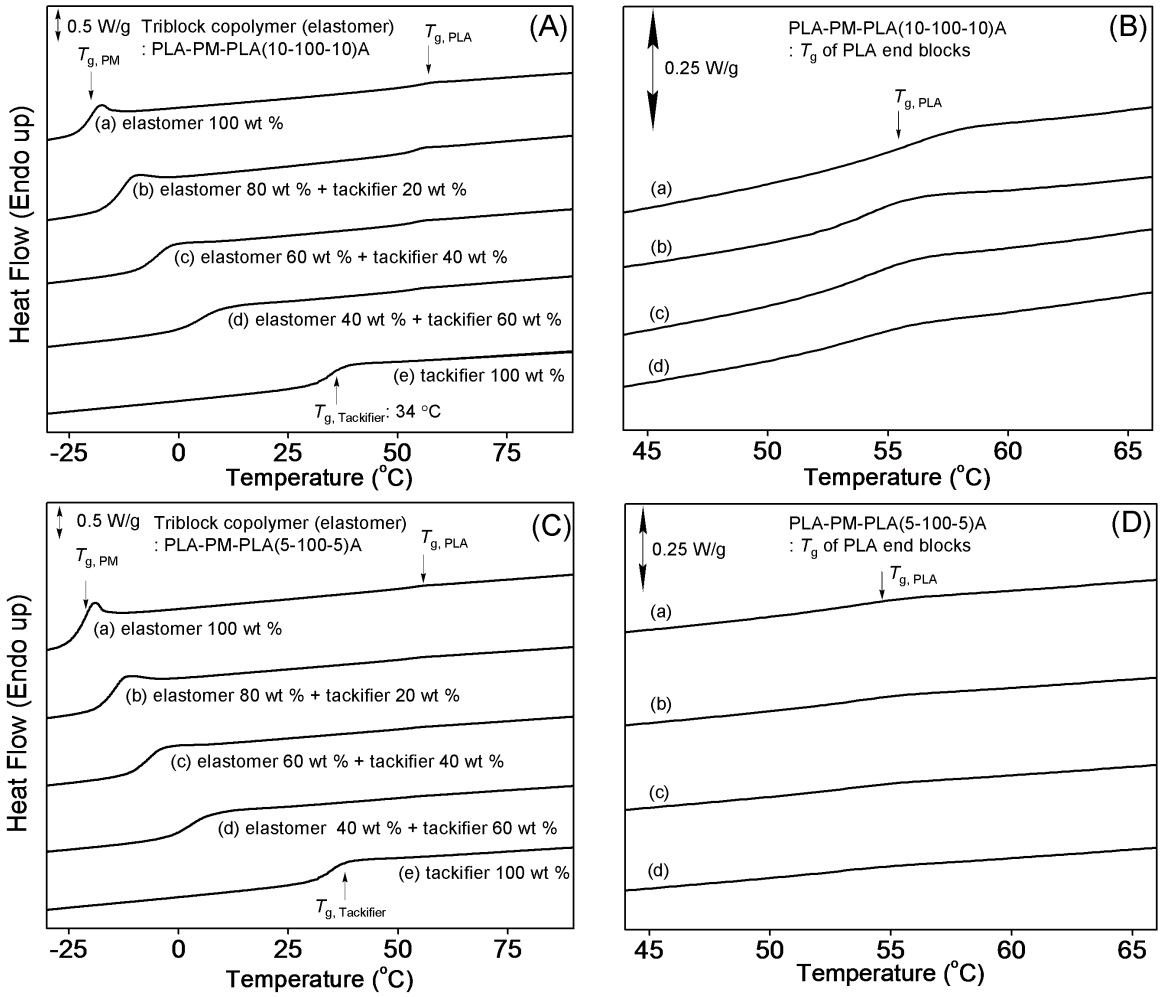

Figure S4. DSC analysis for PLA-PM-PLA(10-100-10)A/RE tackifier blends (A) PLA endblocks in A blends (B) PLA-PM-PLA(5-100-5)A/RE tackifier blends (C) PLA endblocks in C blends (D). Traces represent the second heating after annealing at 120 °C with a heating rate of 10 °C min^{-1}.

Table S1. Adhesive performance data[a]

PSA	180° Peel strength (N/cm)	Tack (N)	Shear strength (min)
Elastomer (5-100-5)A	0.69 ± 0.08	0.52 ± 0.04	2831 ± 529
80 Elastomer (5-100-5)A + 20 tackifier	2.06 ± 0.19	0.69 ± 0.02	1936 ± 697
60 Elastomer (5-100-5)A + 40 tackifier	2.91 ± 0.28	1.01 ± 0.02	2670 ± 615
40 Elastomer (5-100-5)A + 60 tackifier	0.52 ± 0.12[b]	0.91 ± 0.55	1706 ± 517
Elastomer (10-100-10)B	0.40 ± 0.06	0.36 ± 0.03	1479 ± 332
80 Elastomer (10-100-10)B + 20 tackifier	0.64 ± 0.09	0.57 ± 0.04	1936 ± 686
60 Elastomer (10-100-10)B + 40 tackifier	2.88 ± 0.14	1.07 ± 0.09	1248 ± 481
40 Elastomer (10-100-10)B + 60 tackifier	0.36 ± 0.02[b]	0.85 ± 0.43	2623 ± 725
Duct tape (width: 25 mm)	4.20 ± 0.04	0.54 ± 0.09	203 ± 48
Paper tape (width: 25 mm)	2.43 ± 0.08	0.58 ± 0.05	1396 ± 181
Scotch tape (width: 20 mm)	1.94 ± 0.18	0.52 ± 0.04	>10000
Electrical tape (width: 20 mm)	1.87 ± 0.03	0.37 ± 0.02	503 ± 29
Post-it® note (width: 16 mm)	0.33 ± 0.03	0.09 ± 0.03	<0.5

[a] Elastomer is PLA-PM-PLA triblock copolymer [b] The adhesive was separated from backing sheet (PET), which is called ‘cohesive failure’.

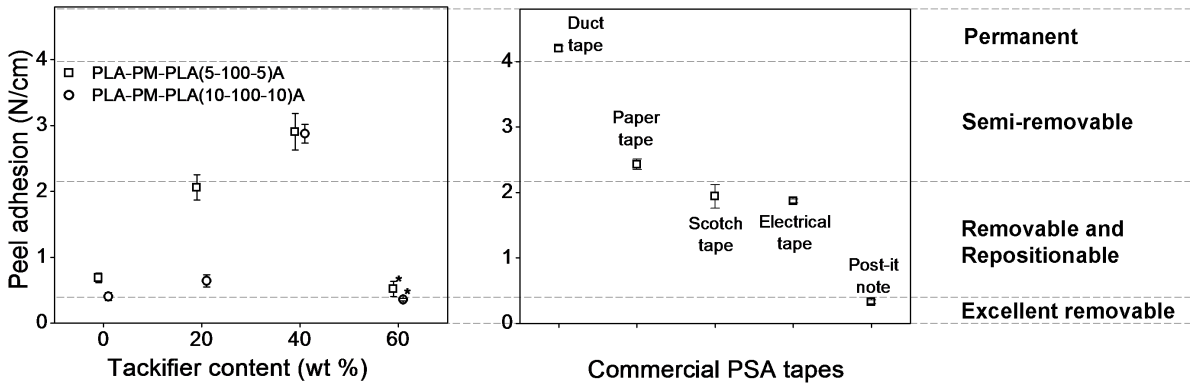

Figure S5. Effect of RE tackifier content on the peel adhesion of the renewable PSA systems (left graph). The peel adhesion of commercial PSA tapes (right graph). The properties of commercial PSAs tapes were classified in the figure. *The adhesive were separated from backing, which is called 'interfacial or adhesive failure'.

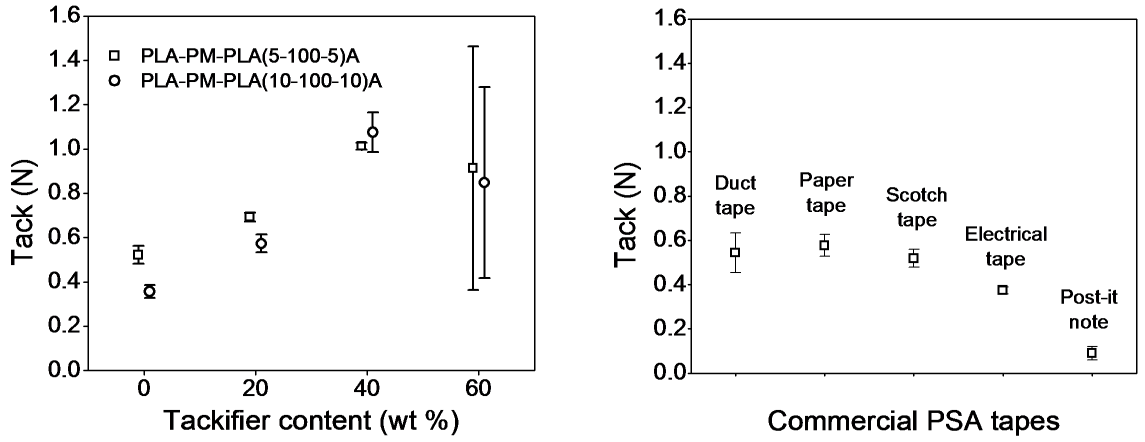

Figure S6. Effect of RE tackifier content on the tack of the renewable PSA systems (left graph). The tack of commercial PSA tapes (right graph). The properties of commercial PSAs tapes were classified in the figure.

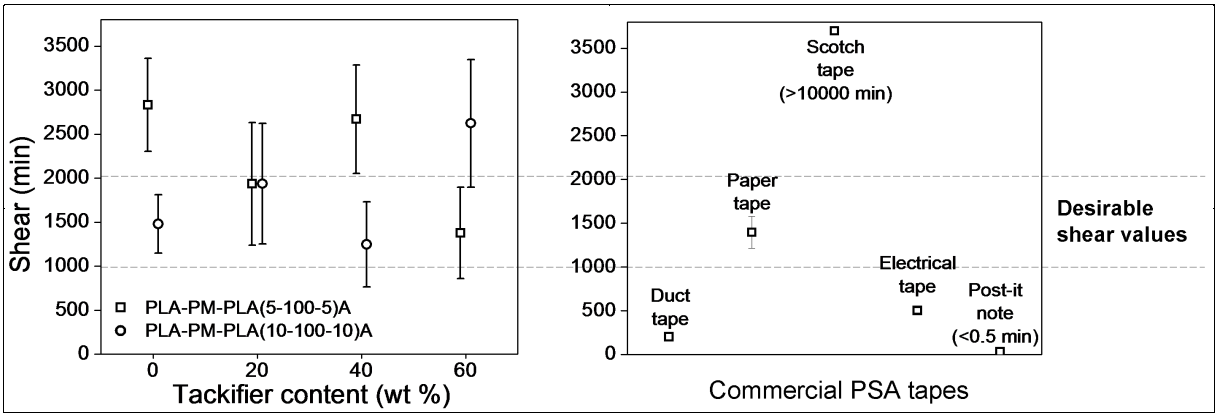

Figure S7. Effect of RE tackifier content on the shear strength of the renewable PSA systems (left graph). The shear strength of commercial PSA tapes (right graph). The desirable shear values of PSAs were indicated in the figure.