Supplementary Information

A Potential Biobased Thermoplastic Elastomer Based on *β*-Myrcene via RAFT-mediated Miniemulsion Polymerization

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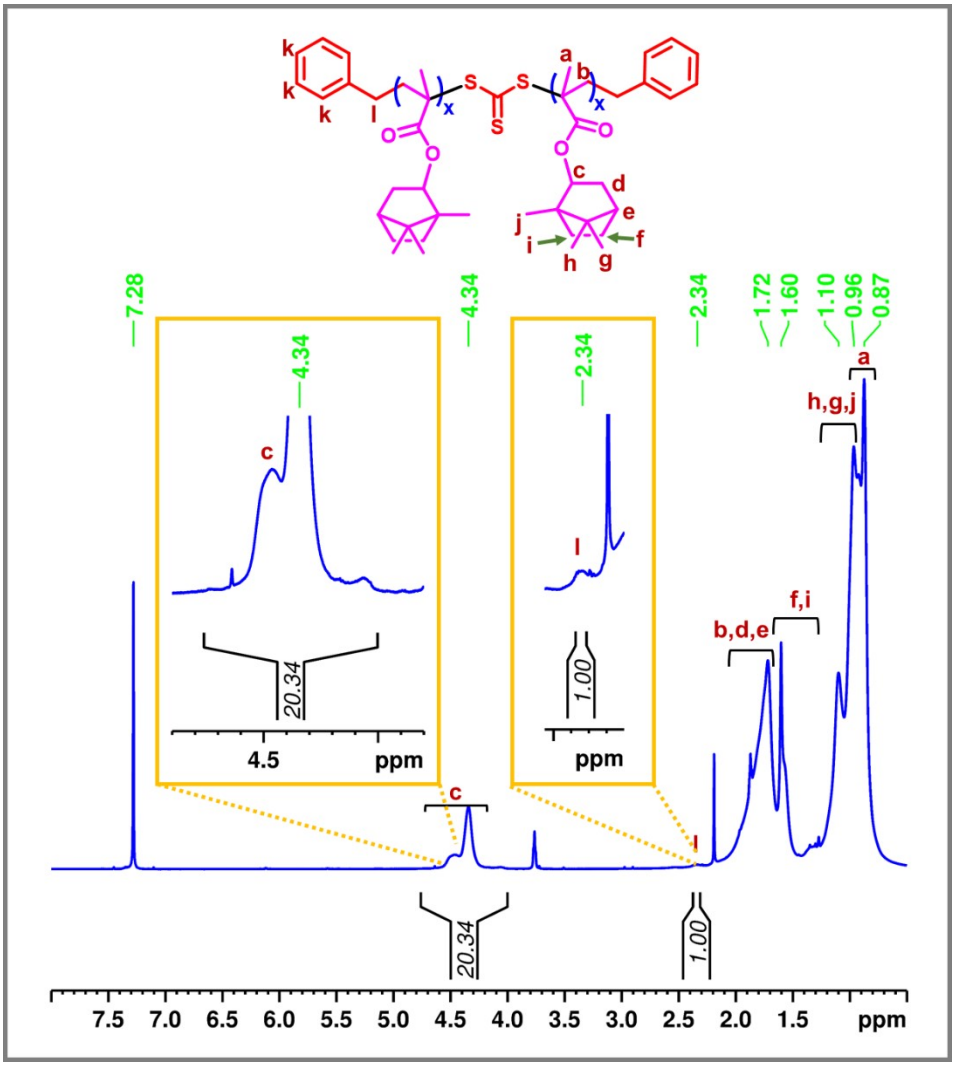

Figure S1: ^{1}H NMR spectrum of PIBMA-CTA-PIBMA macro-RAFT

Figure S1 displays the ^{1}H NMR spectrum of the PIBMA macro-RAFT. The area under the curve for protons "l" and "c" were utilized to compute the molecular weight of the synthesized PIBMA as follows:

$$M.W.\ of\ PIBMA\ = \frac{area\ under\ the\ peak\ for\ "c"\ protons}{area\ under\ the\ peak\ for\ "l"\ protons/2} \times M.W.\ of\ IBMA\ \left(\frac{g}{mol}\right)$$

$$= \frac{20.34}{1/2} \times 222.32\ g/mol$$

$$= (20.34)\ \times 2\ \times 222.32\ g/mol$$

$$\approx 9{,}050\ g/mol$$

This value obtained theoretically is comparable (9,050 g/mol) with that of the molecular weight obtained from GPC analysis (12,000 g/mol).

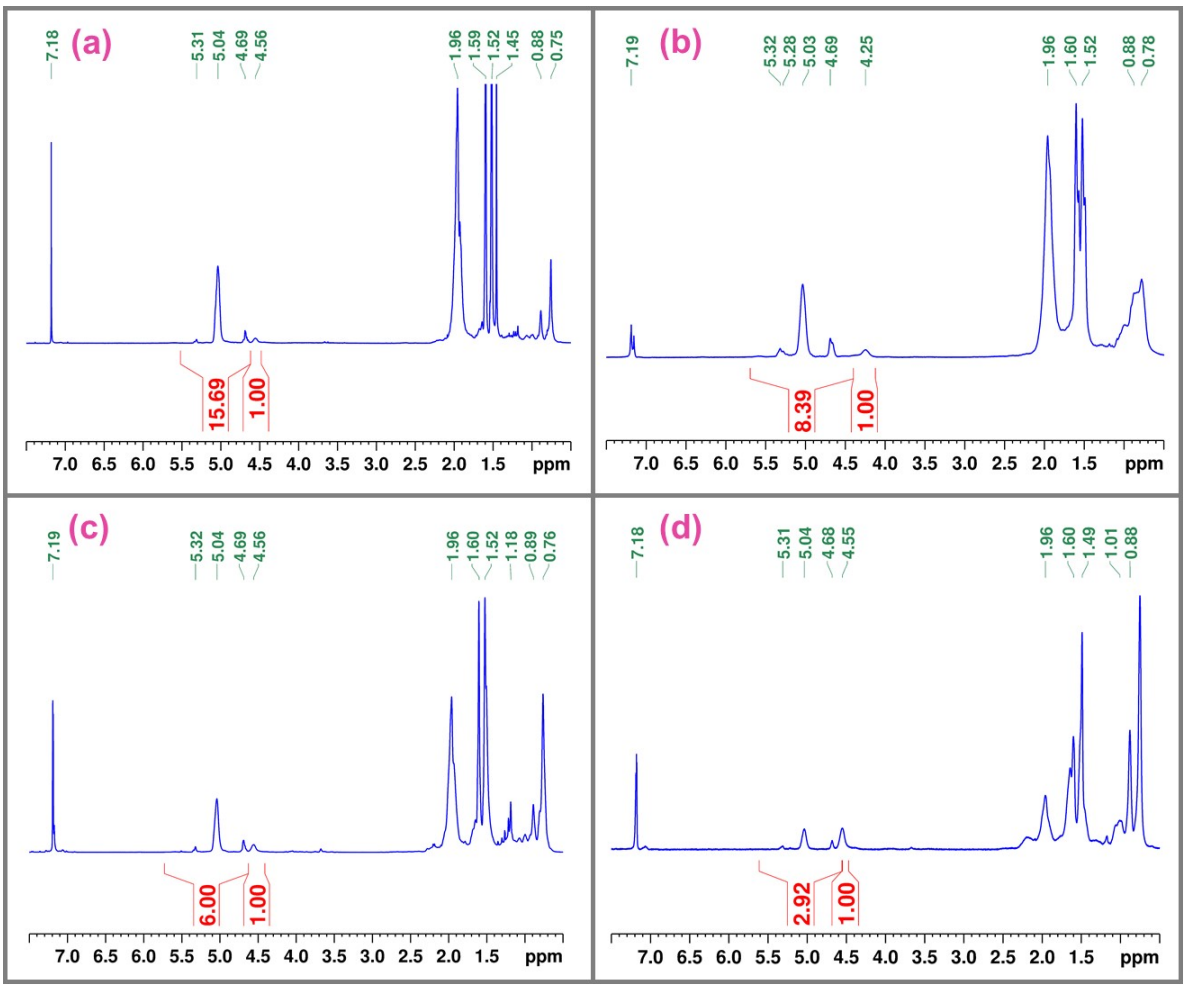

Figure S2: ^{1}H NMR spectra of the synthesized IMI triblock copolymers in $CDCl_3$: (a) IMI-5, (b) IMI-10, (c) IMI-15, and (d) IMI-20 exhibiting the content of IBMA in each of the triBCPs. From ^{1}H NMR spectra, it was observed that the content of IBMA is 5.99, 10.65, 14.29 and 25.51% respectively in IMI-5, IMI-10, IMI-15 and IMI-20 triBCPs, respectively.

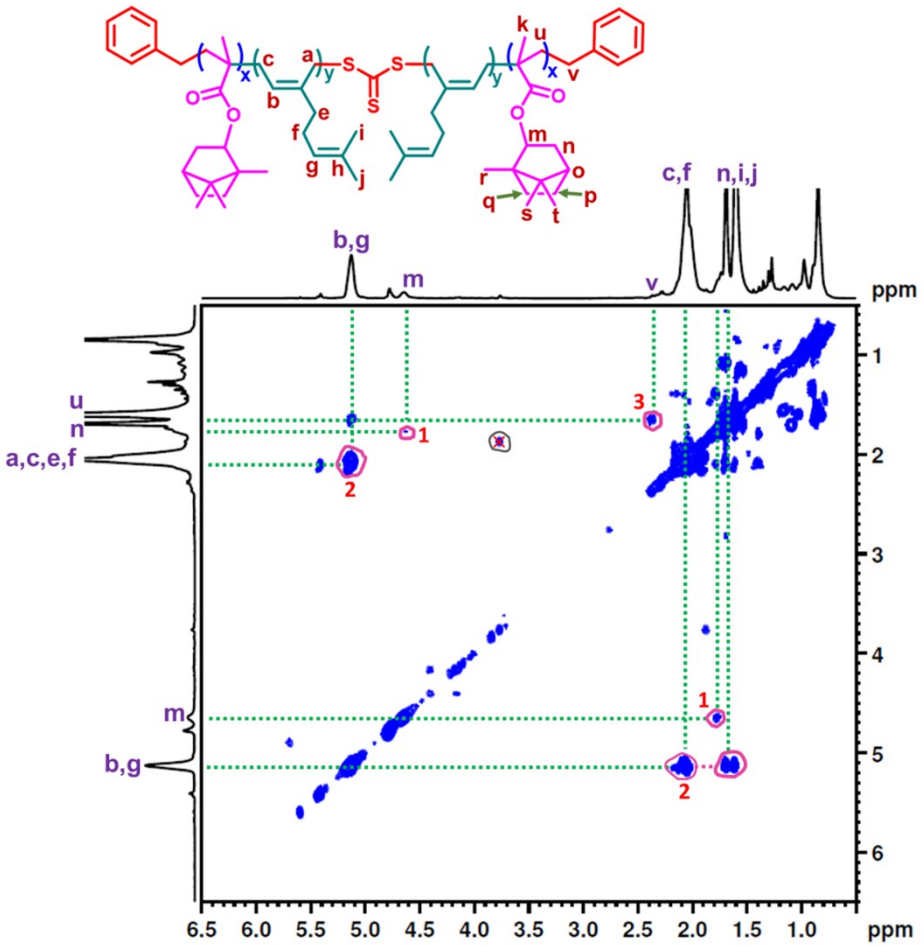

Figure S3: COSY (^{1}H-^{1}H) NMR spectrum in $CDCl_3$ for the synthesized IMI-15 triBCP.

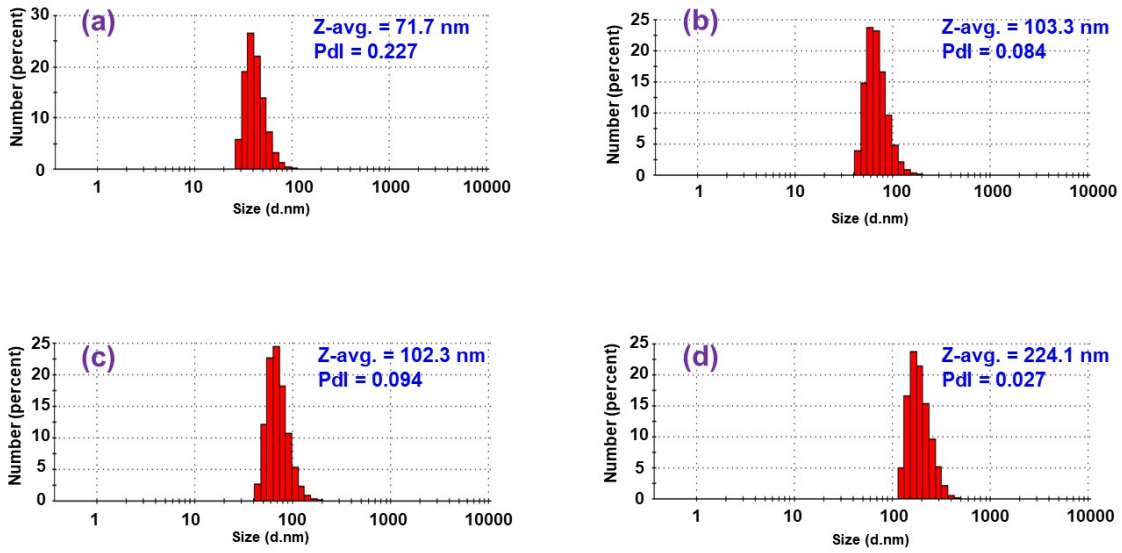

Figure S4: Particle size distribution of the synthesized triblock copolymers latexes obtained using DLS: (a) IMI-20, (b) IMI-15, (c) IMI-10 and (d) IMI-5.

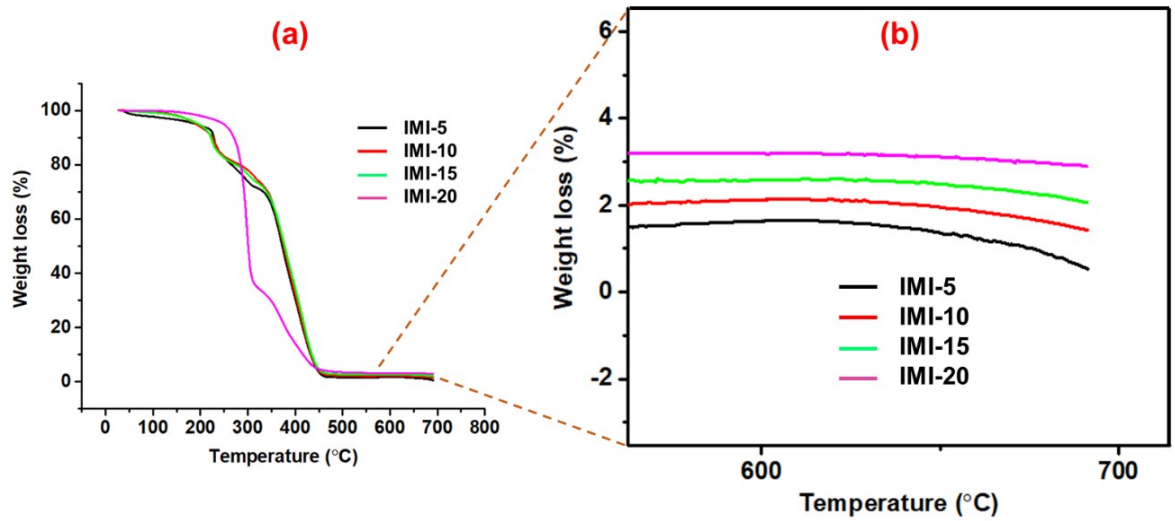

Figure S5: (a) TGA curves of the synthesized IMI copolymers. (b) Zoomed TGA curve for all the copolymers displaying increase in char content for the copolymers with increase in IBMA content.

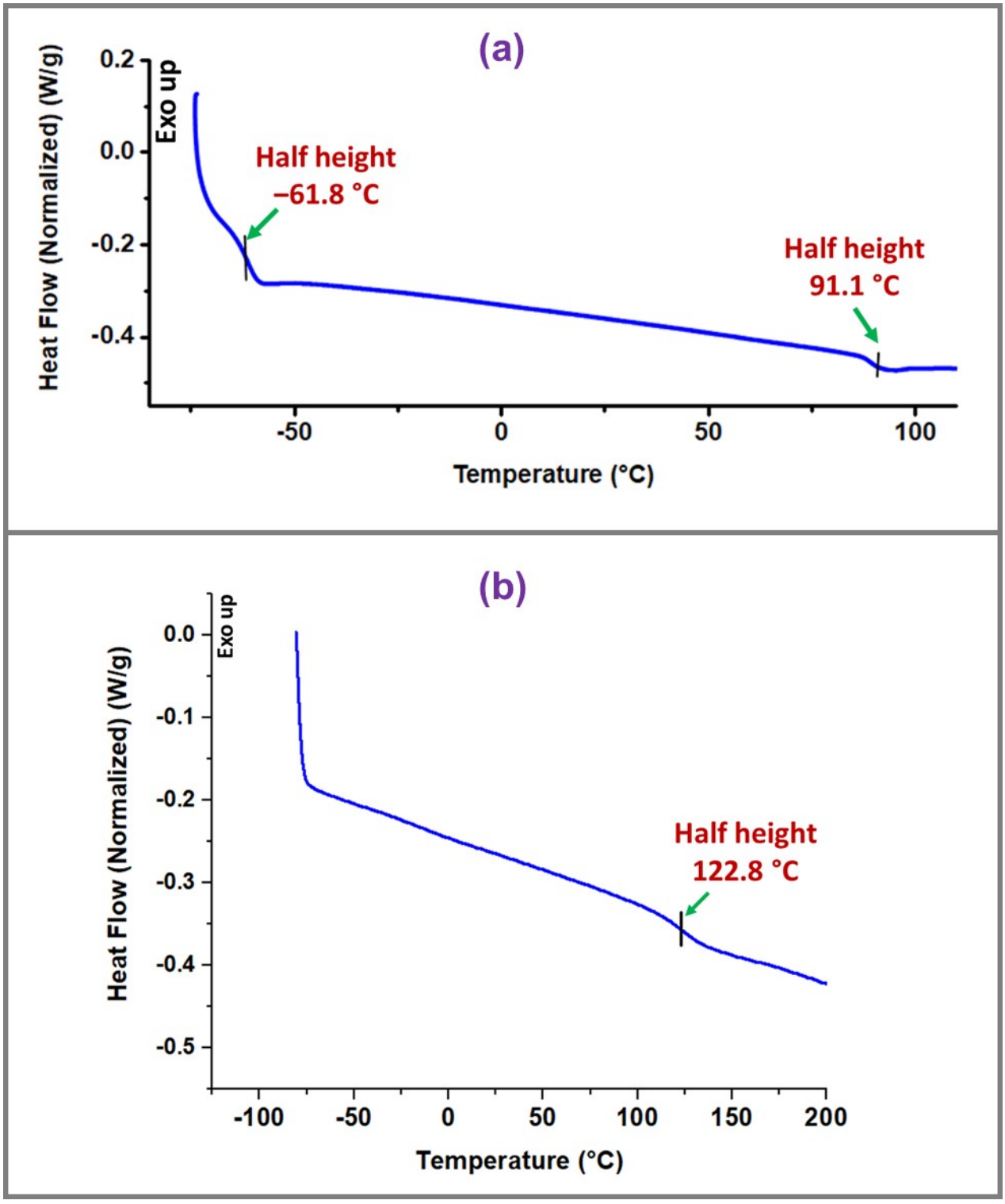

Figure S6: DSC traces of the (a) IMI-15 recorded at 20 °C/min heating rate and (b) PIBMA macro-RAFT (PIBMA1, M_n = 12,000 g/mol)

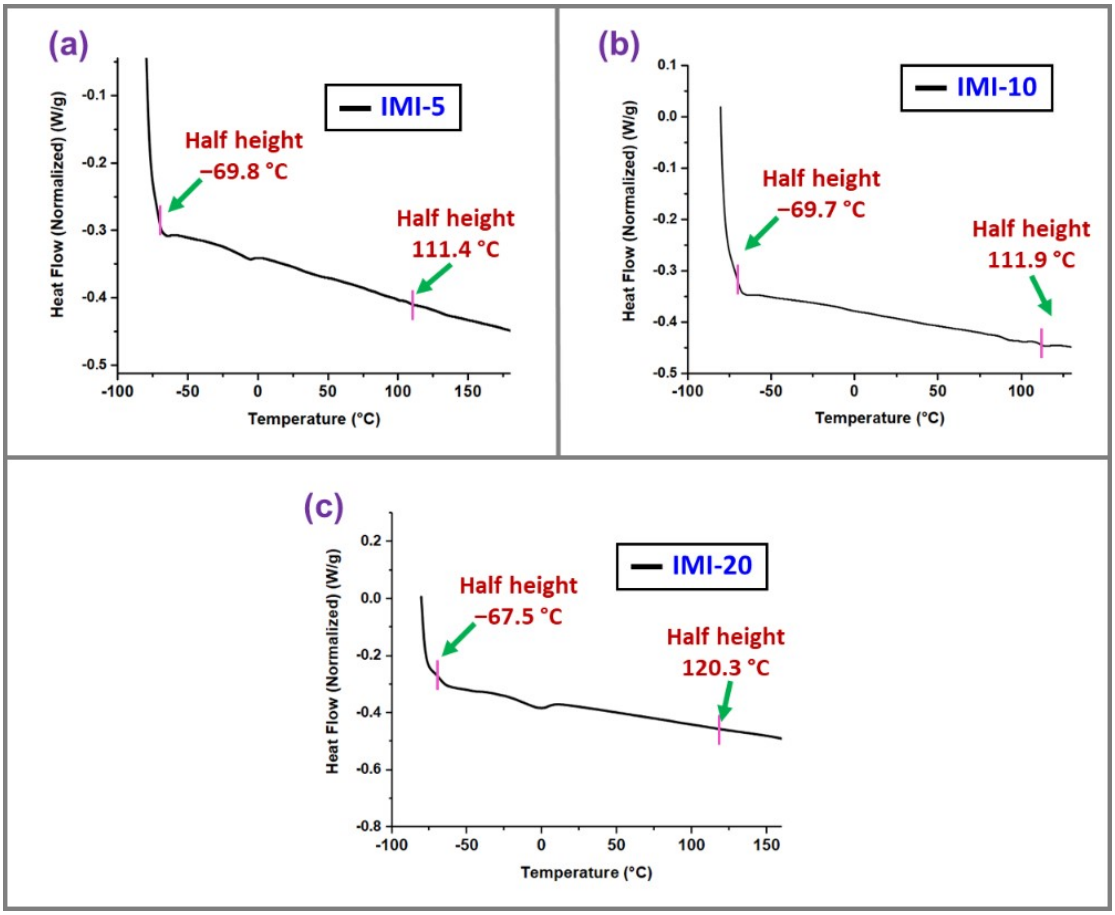

Figure S7: DSC traces of the (a) IMI-5, (b) IMI-10, and (c) IMI-20.

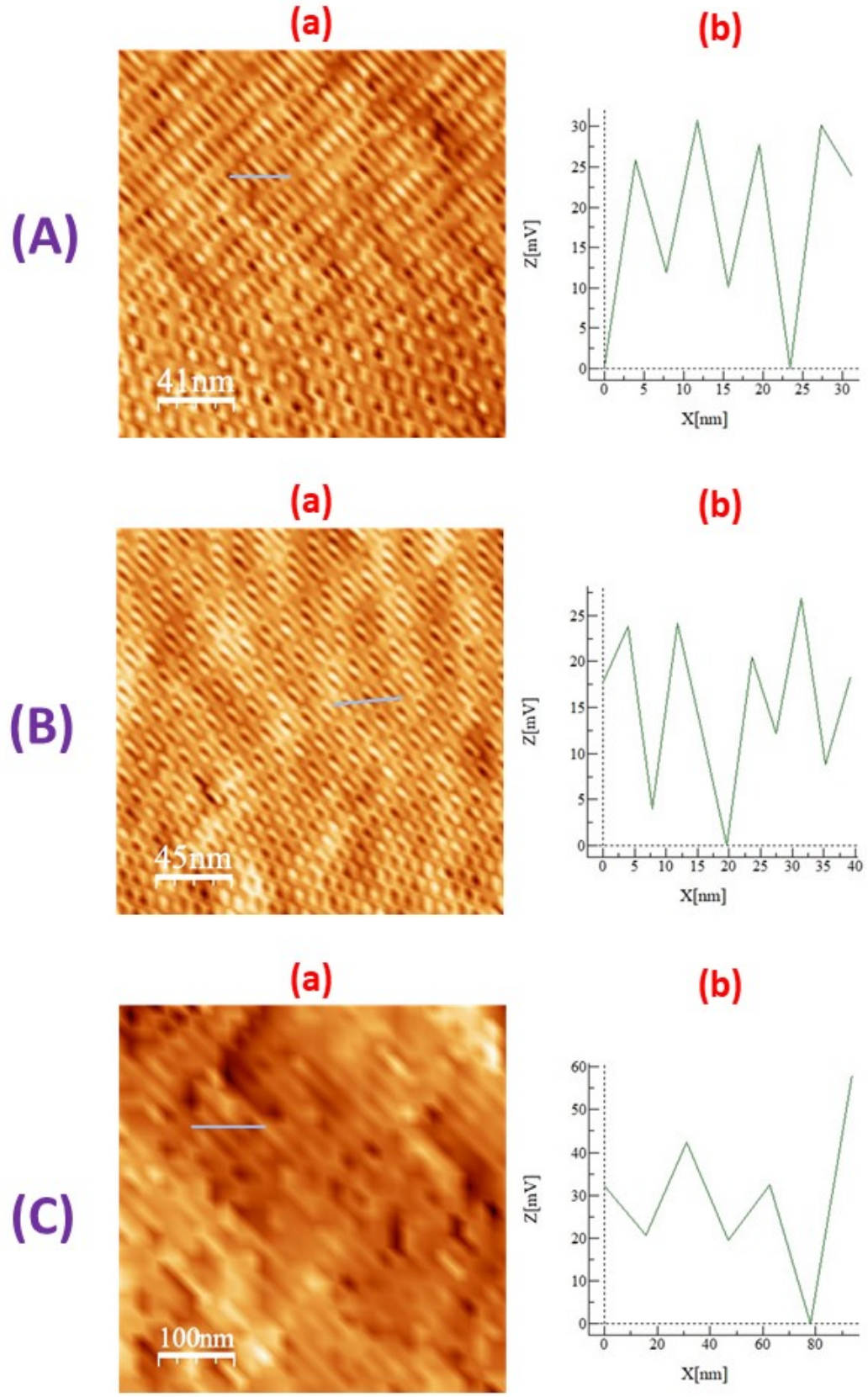

Figure S8: AFM image of the (A) IMI-5, (B) IMI-10, and (C) IMI-15 triBCPs. The respective images correspond to (a) 2D AFM image with a profile mapper, and (b) image profile of the AFM image in relation to the image profile drawn on (b).

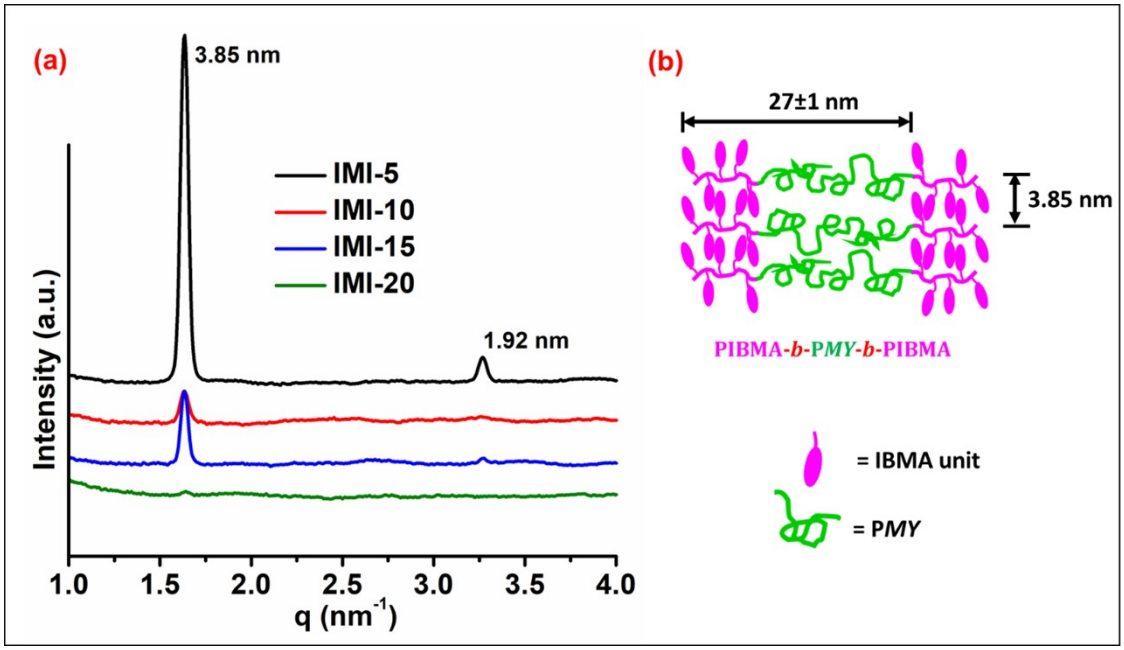

Figure S9: (a) SAXS data of the synthesized IMI triblock copolymers; (b) Schematic representation indicating the microphase separation as well as the organization of IBMA units within the PIBMA blocks based on AFM and SAXS analysis.

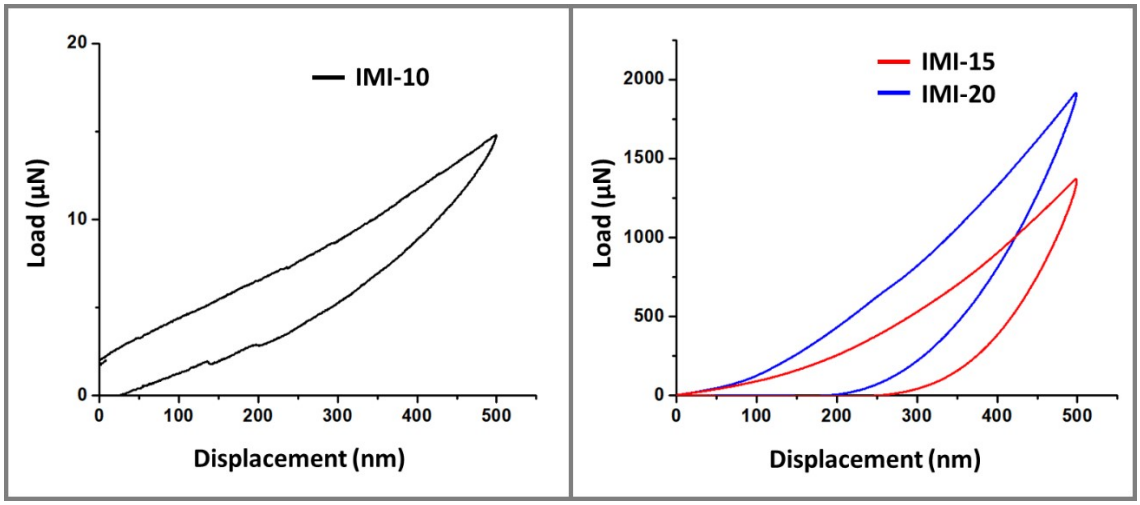

Figure S10: (a) Load vs displacement curve obtained from nanoindentation tests, (b) hardness (H) of the different entries, and (c) modulus (E) of the different entries.

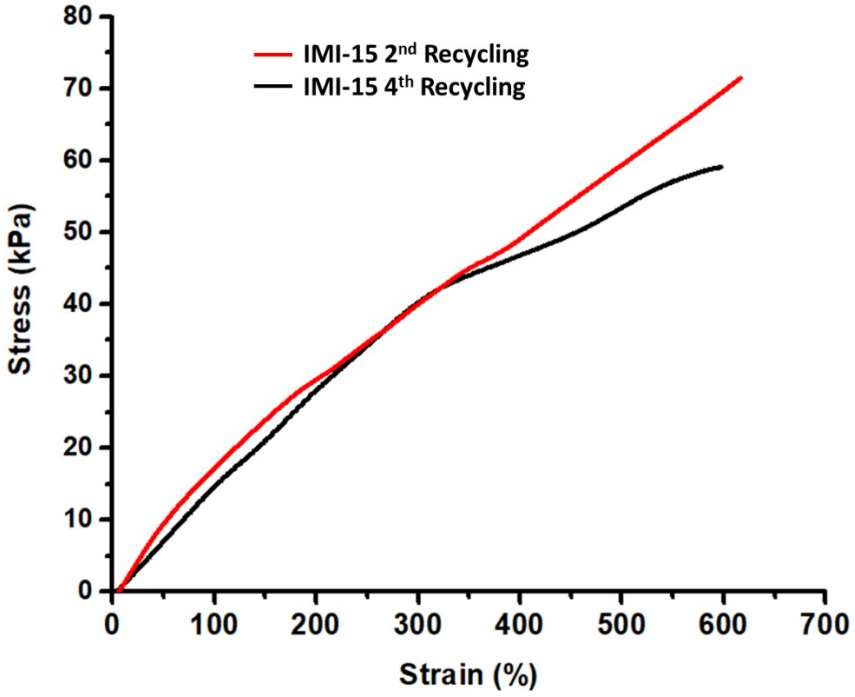

Figure S11: Stress-strain curves of the recycled IMI-15 after 2nd and 4th cycles of recycling. From initial 100 kPa, the final stress became ~72 kPa (2nd recycling) & ~60 kPa (4th recycling), whereas the elongation at break is around 620% (2nd recycling) & 600% (4th recycling) from initial 810% after recycling, exhibiting comparable mechanical properties before and after recycling.

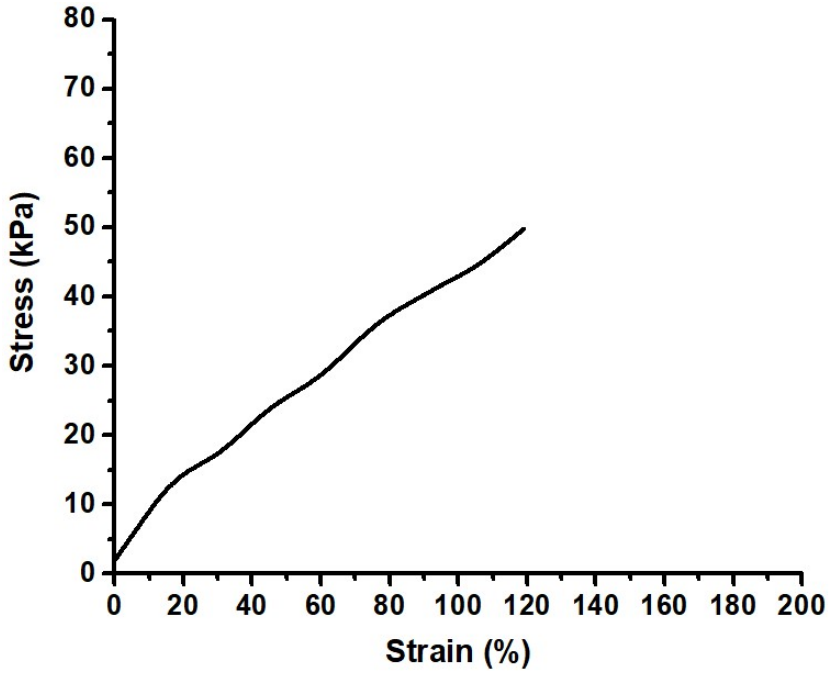

Figure S12: Stress-strain curve of the recycled synthesized IMI-5 after four cycles of recycling. From initial 60 kPa, the final stress became ~50 kPa and the elongation at break is around 110 from initial 105 after four cycles of recycling exhibiting similar mechanical properties before and after recycling.

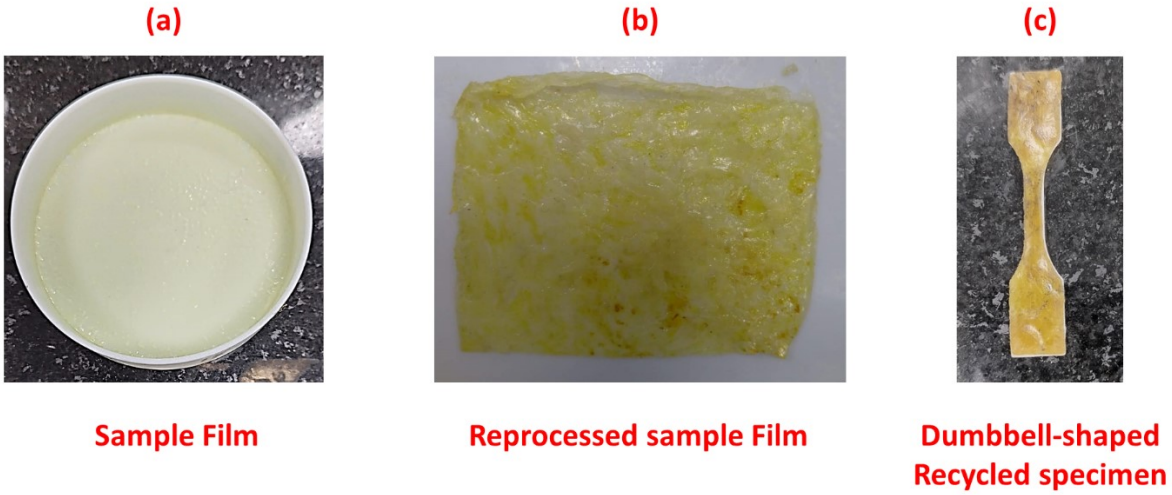

Figure S13: Photographic images of IMI-15: (a) initially casted films on Teflon petri dish; (b) reprocessed film after cut into pieces and remolded at 80 °C and 5 N pressure; (c) dumbbell-shaped specimen of the sample.