Electronic Supplementary Material (ESI) for Polymer Chemistry.

Composition Processing Properties Relationship of Vitrimers Based on Polyethyleneimine

Natanel Jarach[1], Daniel Golani[1], Ofer Asaf[1], Samuel Kenig[1], Hanna Dodiuk[1], Yoav Shamir[2], Amir Goldbourt[2], Naum Naveh[1]*

[1]The Department of Polymer Materials Engineering, Pernick Faculty of Engineering, Shenkar – Engineering. Design. Art. Raman-Gan, Israel.

[2]School of Chemistry, Tel Aviv University, Tel Aviv, Israel.

*Corresponding Author

Table S 1. Gel content, $\overline{M_c}$ and reacted secondary amine groups in PI samples.

Sample	Gel content [%]	$\overline{M_c}\left[\frac{gr}{mol}\right]$	Reacted secondary amine groups [%]
PI	99.4±0.1	240±9	88.0±3.8
PI-10	99.0±0.4	222±2	90.2±0.9
PI-20	99.6±0.1	208±7	97.1±3.7
PI-40	99.5±0.1	204±7	98.5±1.3
PI-60	99.6±0.3	200±1	98.7±0.3
PI-M	98.4±0.7	219±24	89.7±10.2

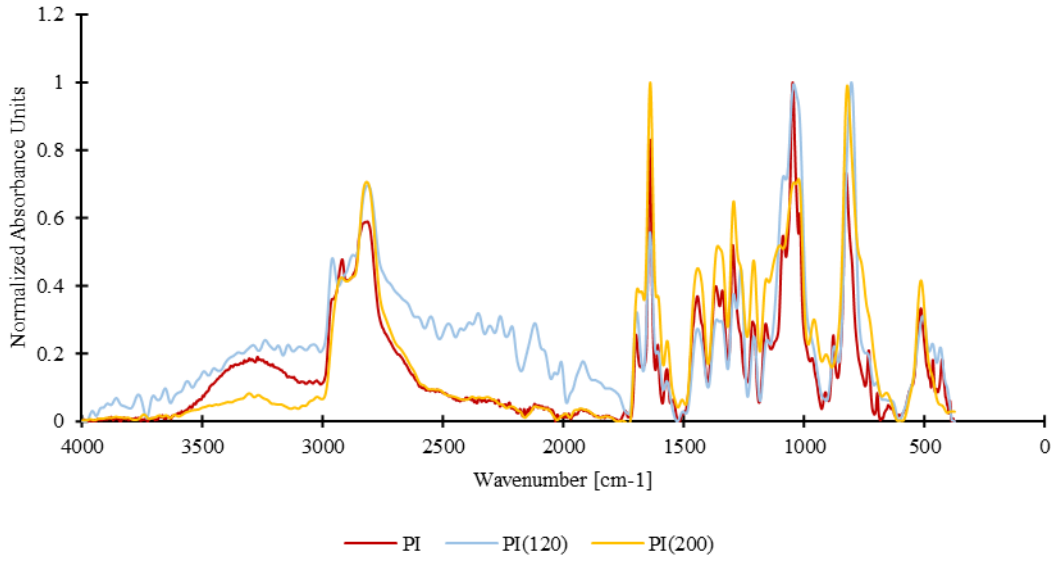


Figure S 1. ATR-IR of PI, PI-120 and PI-200 polyimine samples after drying.

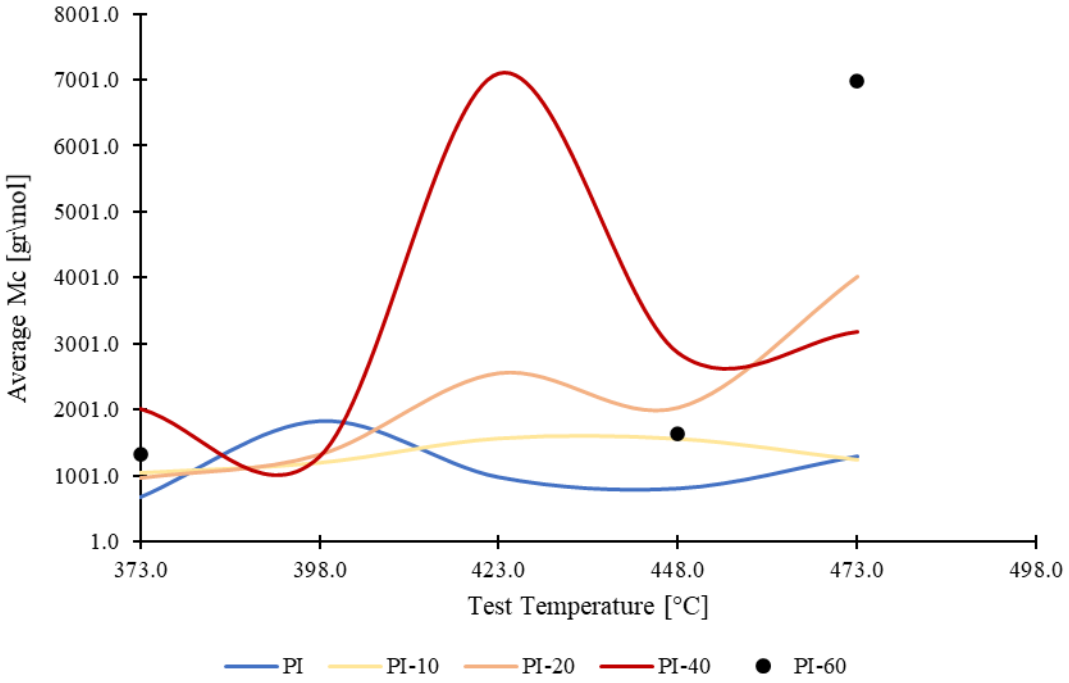
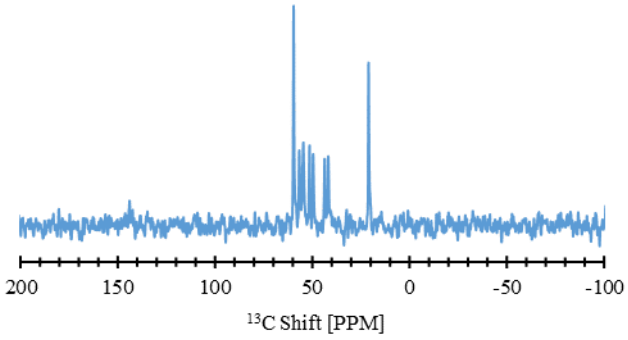


Figure S 2. Cross-linking density of PI, PI-10, PI-20, PI-40 and PI-60 at temperatures based on the rubber elasticity theory.

A)



B)

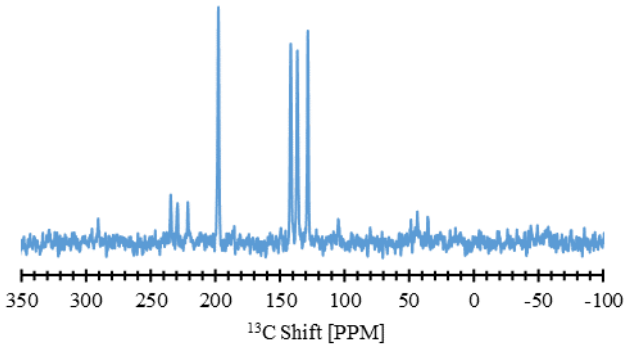
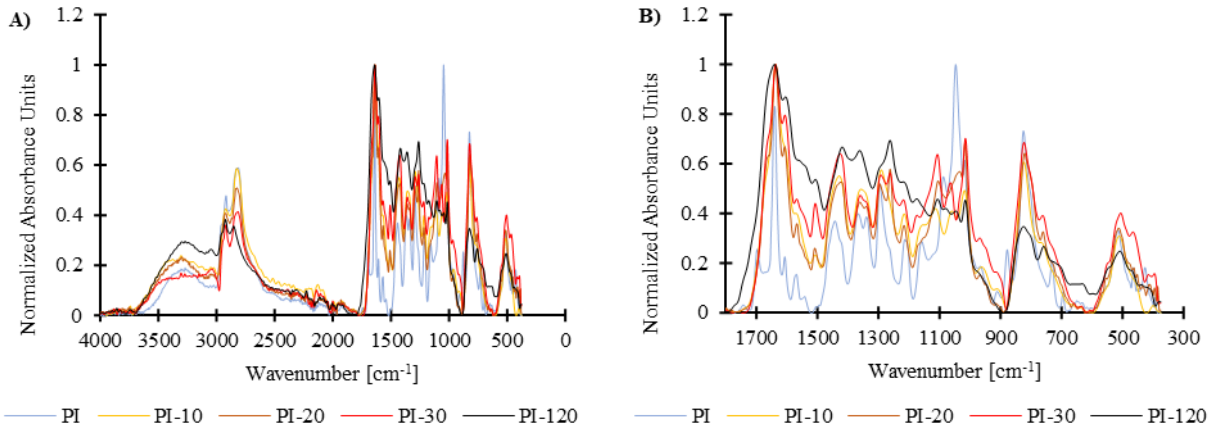


Figure S 3. ^{13}C SS-NMR of PEI (A) and TPA (B) acquired on a 14.1T magnet. PEI spectrum was acquired with a single pulse excitation and low power ^{1}H decoupling using 32 scans and a recycle delay of 13 sec. TPA spectrum was acquired using the CPMAS experiment employing 32 scans and a recycle delay of 20 sec



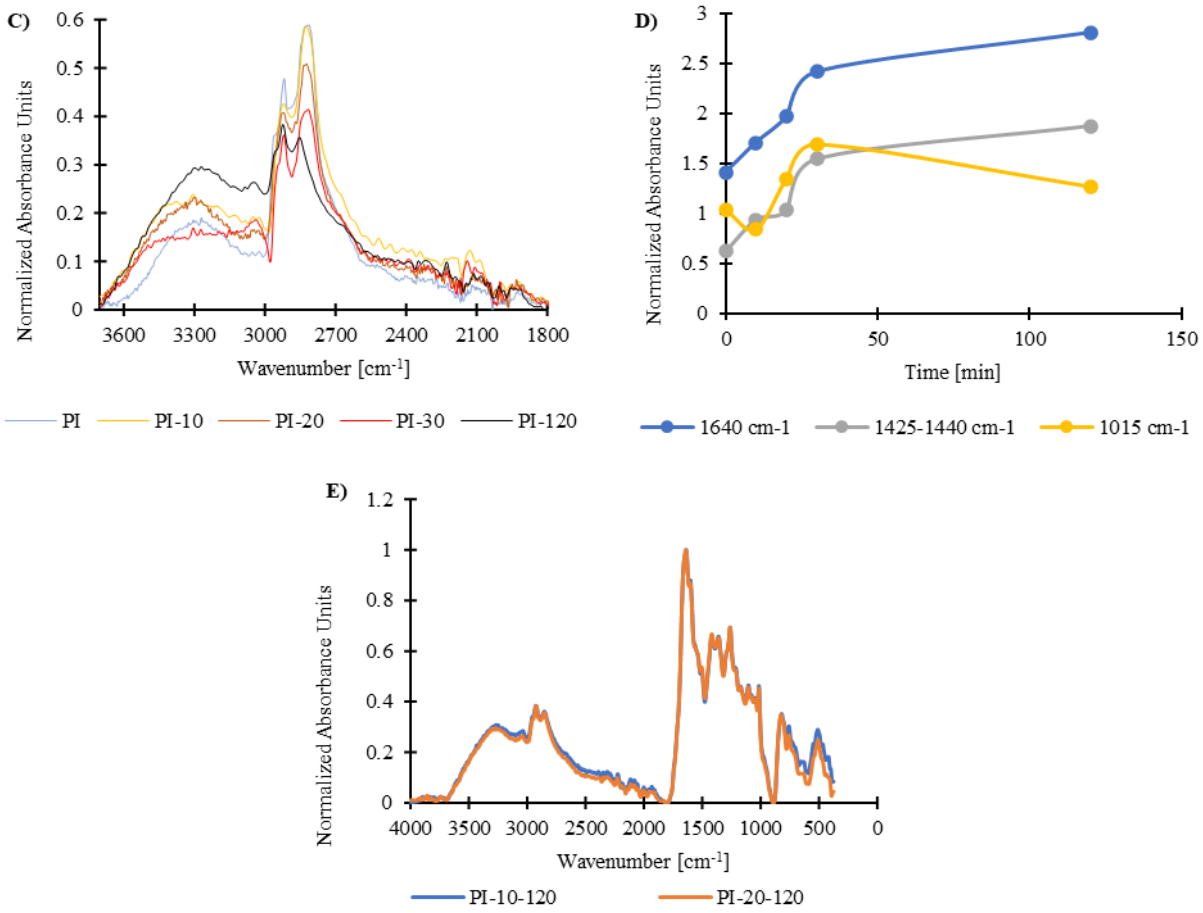


Figure S 4. A – a full range ATR-IR spectra for PI, PI-10, PI-20, PI-30 and PI-120, B – 300-1800 cm-1 ATR-IR spectra for these samples, C – 1800-3700 cm-1 ATR-IR spectra for these samples, D – changing in absorbance for chosen signals (normalized to CH_2 spectra) during time at 220°C, E – ATR-IR spectra for PI-10P and PI-20P.

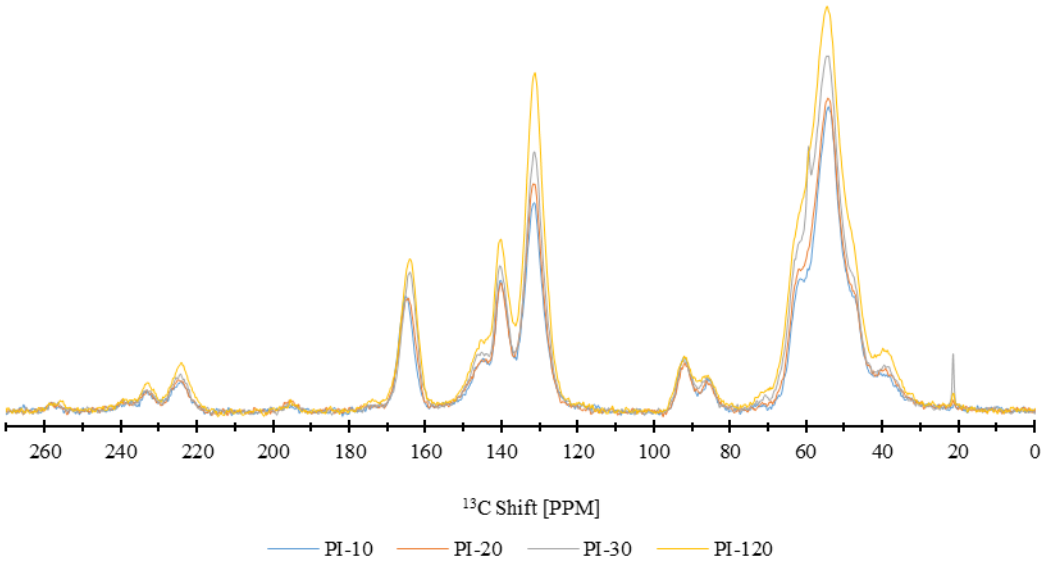


Figure S 5. ^{13}C-CPMAS SS-NMR of PI samples molded at 150°C for 10, 20, 30, and 120 min. All spectra were acquired on a 14.1T magnet using 3000 scans, recycle delays of 4.3-7.7 sec, a CP contact time of 2 ms with 50 kHz ^{13}C power level and 73 kHz ^{1}H power level, and using 80 kHz ^{1}H swf-tppm decoupling.

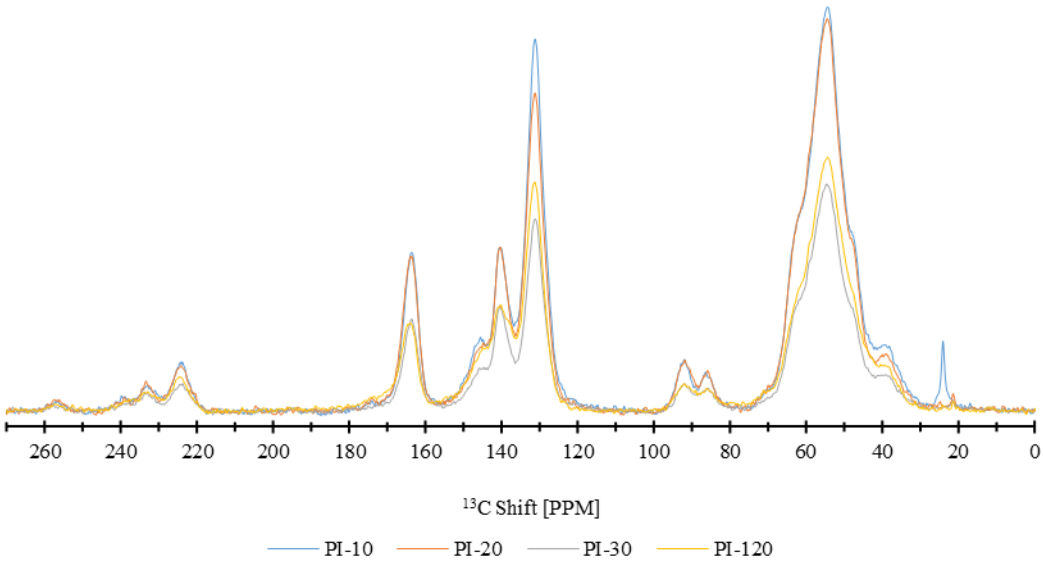


Figure S 6. ^{13}C-CPMAS SS-NMR of PI samples molded at 180°C for 10, 20, 30, and 120 min. All spectra were acquired on a 14.1T magnet using 1600 (PI-30) or 3000 scans and recycle delays of 7-14 sec. Other parameters are similar to Figure S5.

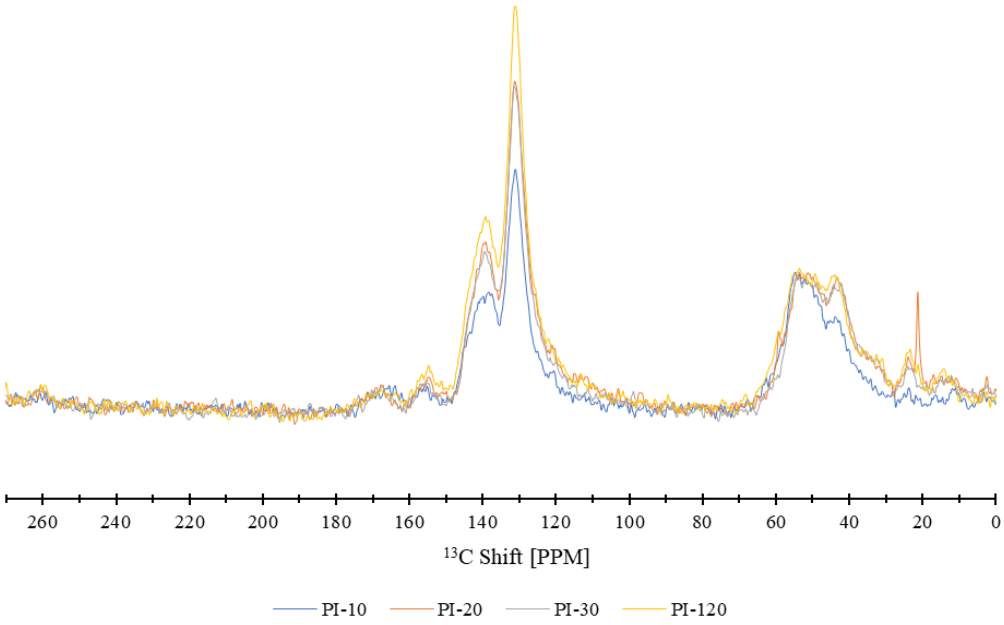


Figure S 7. ^{13}C-multi-CP MAS SS-NMR of PI samples molded at 260℃ for 10, 20, 30, and 120 min. All spectra were acquired on a 9.4T magnet using 5500 scans, recycle delays of 3-4 sec, a CP contact time of 2 ms with 50 kHz ^{13}C power level and 86 kHz ^{1}H power level, 11 CP repetitions with a repolarization time of 0.2 sec, and using 100 kHz ^{1}H swf-tppm decoupling.

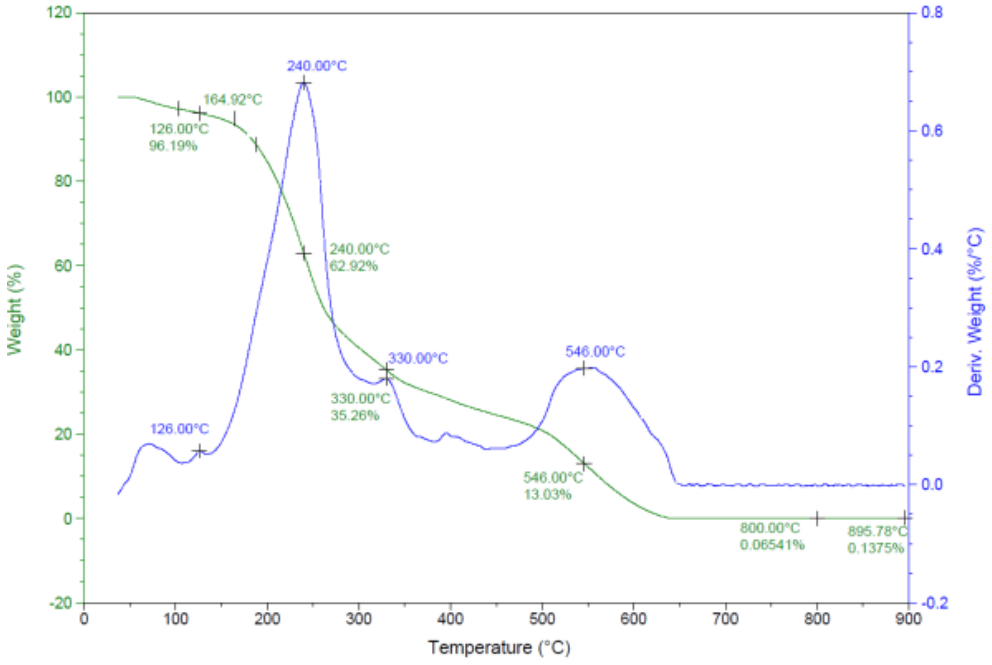


Figure S 8. TGA results of PEI.

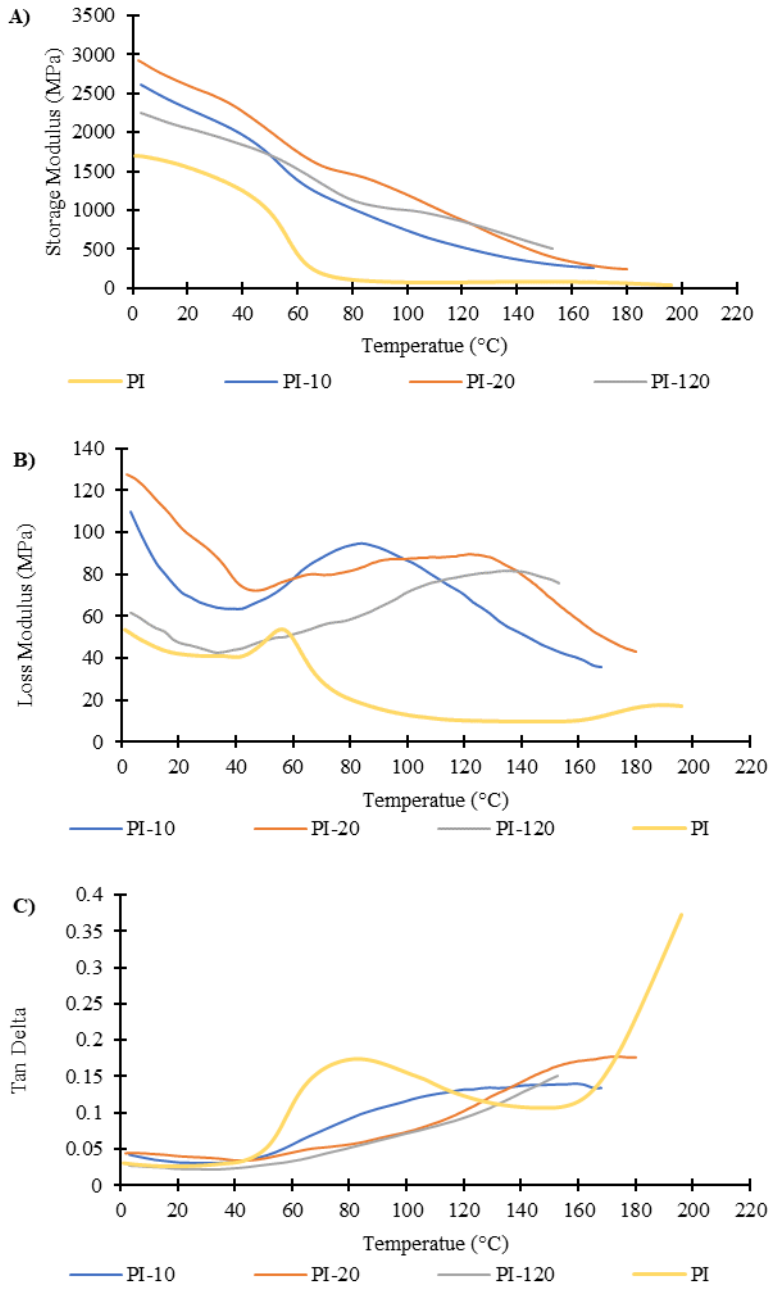


Figure S 9. Dynamic flexural results (DMA) – storage modulus (A), loss modulus (B) and tan delta (C).