

Supporting Information

Assembling Conductive PEBA Copolymer at the Continuous Interface in Ternary Polymer Systems: Morphology and Resistivity

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1. Rheology

The rheological properties of the raw materials were examined on a MCR 301 rheometer (Anton Paar) with a parallel-plate configuration. A gap of 1 mm was used in all cases. Strain sweeps were first performed to determine the linear viscoelastic region. The thermal stability was evaluated in the oscillation mode with a frequency of 1 Hz at 200°C or 250°C for 20 min. The change of complex viscosity was found to be within 2% for all the materials at 200°C. At 250°C, the complex viscosities of PEBA and PET decreased by 17% and 11% respectively; the changes for LDPE and PS were within 1%. The frequency sweep was then performed for all the samples at a strain of 5–10%. N₂ was used all the time when applicable to minimize the degradation.

The complex viscosities of the neat polymers at the temperature corresponding to the processing temperature (200°C and 250°C) were examined and the results are shown in Fig. S1. At 50 RPM in the Brabender mixer, the shear rate is estimated to be around 25 s⁻¹.¹ Under these conditions, the viscosity of PEBA is much lower than that of the other polymers. The viscosities of the host polymers (components in addition to PEBA) are very close.

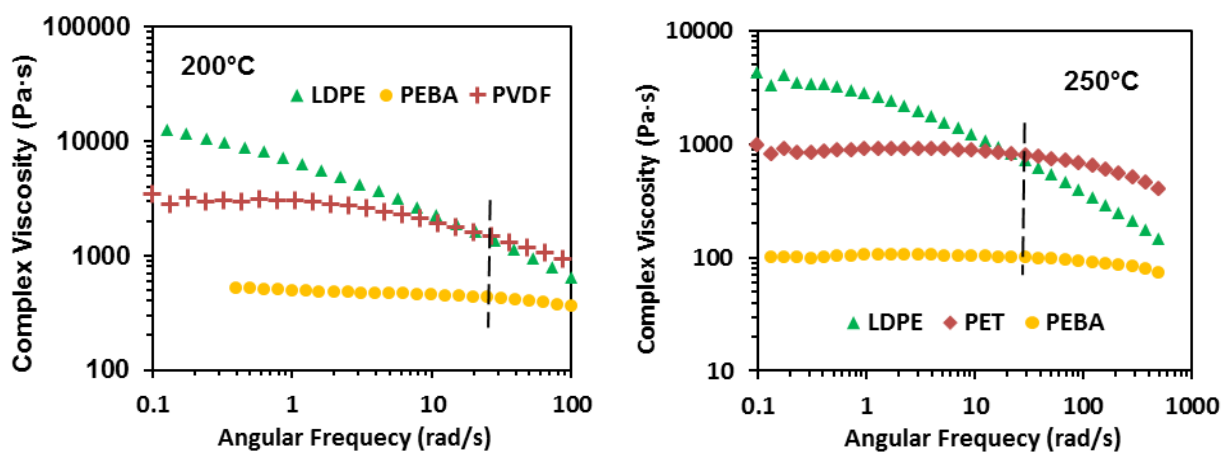


Figure S1. Complex viscosities of the neat polymers at 200 and 250°C. The dashed line indicates the processing shear rate of 25 s⁻¹.

2. Interface Coverage

The interface coverage by PEBA at the LDPE/PET interface in LDPE/PEBA/PET is estimated by the following equation:

$$\text{Interface coverage (\%)} = \frac{A_1}{A_0} \times 100$$

where A_0 is the area circled by the red line (area of LDPE/PET interface) and A_1 is the total area occupied by PEBA within the circled region (Fig. S2). Fig. S3 shows the evolution of the interface coverage as a function of PEBA composition in the LDPE/PEBA/PET blends. A_0 and A_1 were obtained by using a digitizing table from Wacom and SigmaScan v.5 software. In this method, the interface coverage in real three-dimensional space is approximated by its projection on the two-dimensional SEM image.

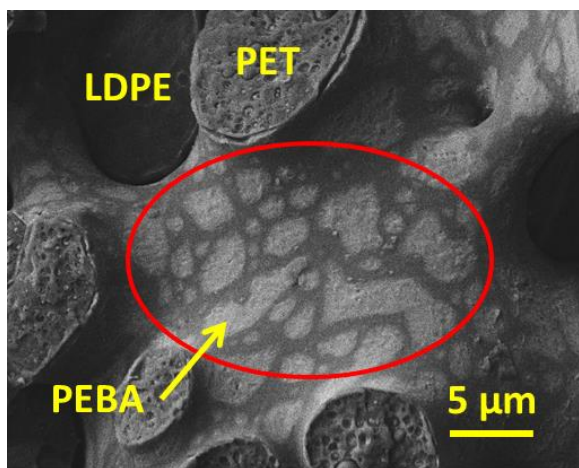


Figure S2. An example of an LDPE/PEBA/PET cryo-fractured sample for interface coverage calculation

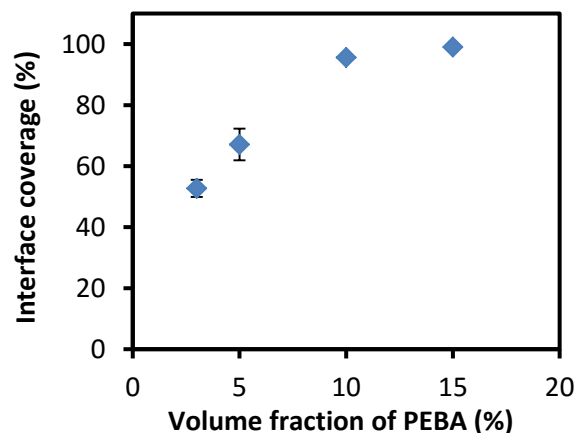


Figure S3. Interface coverage by PEBA at the LDPE/PET interface in LDPE/PEBA/PET blends

3. Volume Resistivity of the Blends

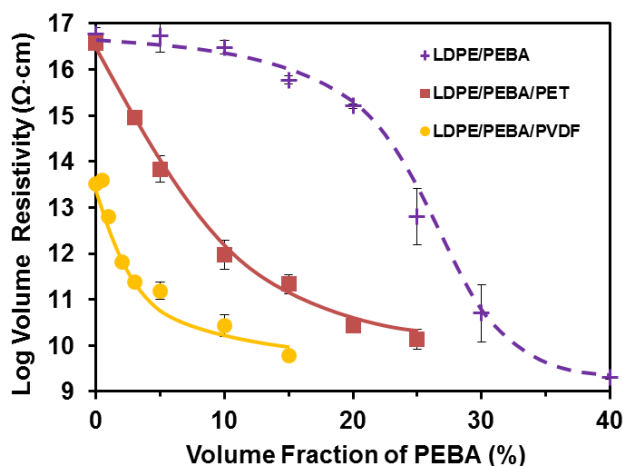


Figure S4. Volume resistivity of the blends.

Note that the initial lower volume resistivity for the LDPE/PEBA/PVDF system at 0% PEBA is due to the significant low volume resistivity of neat PVDF ($10^{13} \Omega\cdot\text{cm}$) as compared to other host polymers ($10^{16} - 10^{17} \Omega\cdot\text{cm}$) (see Table 1 in the manuscript).

4. In-situ Measurement of the Neumann Angle for LDPE/PEBA/PET^{2,3}

The blend of LDPE/PEBA/PET (50/3/47) processed in Brabender internal mixer was subjected to annealing at 260°C for 10 min. The sample was cryo-microtomed and stained by phosphotungstic acid. The SEM images were then taken after gold coating. In order to determine the Neumann angle θ , the LDPE/PEBA and PET/PEBA interfaces were first fitted by complete circles (Fig. S5). The symmetrical axis of the two circles is defined by their centers (O1 and O2). The two centers are joined to the line of 3-phase contact respectively by the segments PO1 and PO2. The sum of the formed angles α and β mathematically equals the Neumann angle θ (defined by the tangent lines at P of the two circles) as shown in Fig. S5. The Neumann angle was then averaged based on 7 measurements: $\theta = 86^\circ \pm 8^\circ = 1.5 \pm 0.1$ (in radians). The result

corresponds very well with that obtained by Eqn. (1) from the interfacial tension values in the manuscript.

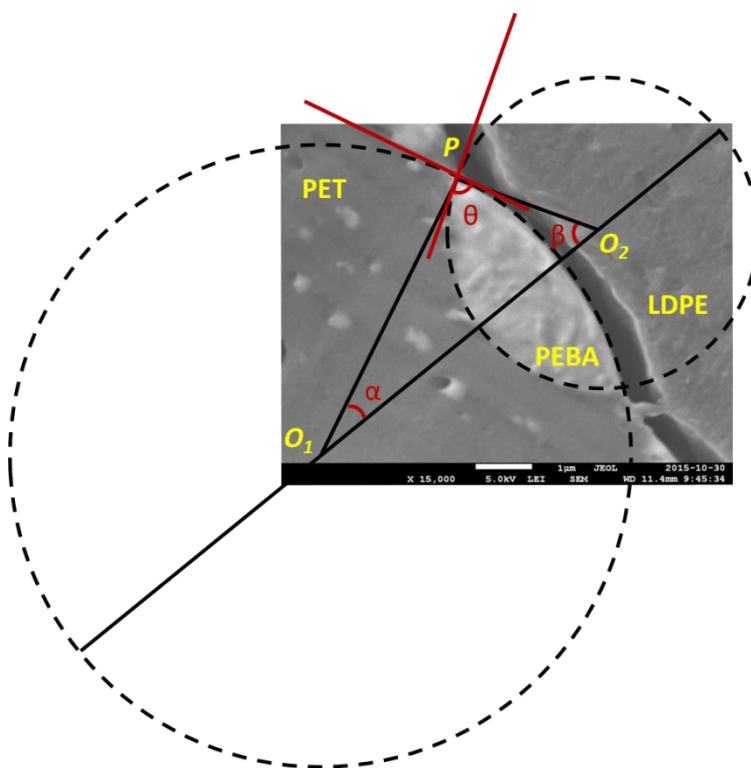


Figure S5. Geometrical parameters of the LDPE/PEBA/PET (50/3/47) blend for determining the Neumann angle θ .

4. References

- (1) Bousmina, M.; Ait-Kadi, A.; Faisant, J. B. Determination of Shear Rate and Viscosity from Batch Mixer Data. *J. Rheol.* **1999**, 43, (2), 415-433.
- (2) Torza, S.; Mason, S. G. Three-phase Interactions in Shear and Electrical Fields. *J. Colloid Interface Sci.* **1970**, 33, (1), 67-83.
- (3) Virgilio, N.; Desjardins, P.; L'Espérance, G.; Favis, B. D. In Situ Measure of Interfacial Tensions in Ternary and Quaternary Immiscible Polymer Blends Demonstrating Partial Wetting. *Macromolecules* **2009**, 42, (19), 7518-7529.