Supporting information

Morphology Mapping of Nanoparticle-filled Immiscible Polymer Blends in Flow: The Existence of a Critical Ratio between Nanoparticle Concentration and Droplet Concentration

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Initial morphology of hydrophilic silica filled PS-minor blends



Figure S1 Initial morphology of hydrophilic silica nanoparticle (A200) filled PP/PS blends with **PP as a matrix**: (a-d) PP/PS 95/5 (PS5) blends, (e-h) PP/PS 90/10 (PS10) blends and (i-l) PP/PS 80/20 (PS20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of A200 (A1, A3 and A5) nanoparticles, respectively.

Initial morphology of hydrophobic silica filled PS-minor blends



Figure S2 Initial morphology of hydrophobic silica nanoparticle (R974) filled PP/PS blends with **PP as a matrix**: (a-d) PP/PS 95/5 (PS5) blends, (e-h) PP/PS 90/10 (PS10) blends and (i-l) PP/PS 80/20 (PS20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of R974 (R1, R3 and R5) nanoparticles, respectively.

Initial morphology of hydrophilic silica filled PP-minor blends



Figure S3 Initial morphology of hydrophilic silica nanoparticle (A200) filled PP/PS blends with **PS as a matrix**: (a-d) PP/PS 5/95 (PP5) blends, (e-h) PP/PS 10/90 (PP10) blends and (i-l) PP/PS 20/80 (PP20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of A200 (A1, A3 and A5) nanoparticles, respectively.

Initial morphology of hydrophobic silica filled PP-minor blends



Figure S4 Initial morphology of hydrophobic silica nanoparticle (R974) filled PP/PS blends with **PS as a matrix**: (a-d) PP/PS 5/95 (PP5) blends, (e-h) PP/PS 10/90 (PP10) blends and (i-l) PP/PS 20/80 (PP20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of R974 (R1, R3 and R5) nanoparticles, respectively.

Viscoelastic properties of nanoparticle filled PP-minor blends



Figure S5 Rheological properties of nanoparticle filled PP/PS blends with **PS as a matrix**: (a-b) PP/PS 5/95 (PP5) blends, (c-d) PP/PS 10/90 (PP10) blends and (e-f) PP/PS 20/80 (PP20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of A200 hydrophilic (A1, A3 and A5) and R974 hydrophobic (R1, R3 and R5) silica nanoparticles, respectively.



Viscoelastic properties of nanoparticle filled PP-minor blends

Figure S6 Rheological properties of nanoparticle filled PP/PS blends with **PP as a matrix**: (a-b) PP/PS 95/5 (PS5) blends, (c-d) PP/PS 90/10 (PS10) blends and (e-f) PP/PS 80/20 (PS20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of A200 hydrophilic (A1, A3 and A5) and R974 hydrophobic (R1, R3 and R5) silica nanoparticles, respectively.

Morphology of hydrophilic silica filled PS-minor blends after shearing



Figure S7 Optical micrographs of hydrophilic silica nanoparticle (A200) filled PP/PS blends with PP as a

matrix: (a-e) PP/PS 95/5 (PS5) blends, (f-j) PP/PS 90/10 (PS10) blends and (k-o) PP/PS 80/20 (PS20) blends unfilled and filled with different concentrations (0.5, 1, 3 and 5 wt.%) of A200 (A0.5, A1, A3 and A5) nanoparticles after shearing at 0.1 s⁻¹ and 200 °C for 7000 s, respectively. For these blends, the ratio of A200 NP concentration to PS concentration is varied from 0.05 to 1.0. The scale bar denotes 50 μ m for all images. The red arrow represents the direction of shear flow.

Morphology of hydrophilic silica filled PP-minor blends after shearing



Figure S8 Optical micrographs of hydrophilic silica nanoparticle (A200) filled PP/PS blends with PS as a matrix: (a-d) PP/PS 5/95 (PP5) blends, (e-h) PP/PS 10/90 (PP10) blends and (i-l) PP/PS 20/80 (PP20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of A200 (A1, A3 and A5) nanoparticles after shearing at 0.1 s⁻¹ and 200 °C for 7000 s, respectively. The scale bar denotes 50 μ m for all images. The red

arrow represents the direction of shear flow.

Morphology of hydrophobic silica filled PS-minor blends after shearing

(a) PS5 50µm	(g) PS10.	(m) PS20
(b) PS5-R1	(h) PS10-R0.5	(n) PS20-R1
(c) PS5-R1.5	(i) PS10-R1	(o) PS20-R3
(d) PS5-R2	(j) PS10+R3	(p) PS20-R5
(e) PS5-R3	(k) PS10-R4	(q) PS20-R6
(f) PS5-R5	(I) PS10-R5	(r) PS20-R8

Figure S9 Optical micrographs of hydrophobic silica nanoparticle (R974) filled PP/PS blends with PP as a matrix: (a-f) PP/PS 95/5 (PS5) blends, (g-l) PP/PS 90/10 (PS10) blends and (m-r) PP/PS 80/20 (PS20) blends unfilled and filled with different concentrations (0.5, 1, 1.5, 3, 4, 6 and 8 wt.%) of R974 (R0.5, R1, R1.5, R3, R4, R6 and R8) nanoparticles after shearing at 0.1 s⁻¹ and 200 °C for 7000 s, respectively in which the ratio of R974 NP concentration to PS concentration is varied from 0.05 to 1.0. The scale bar denotes 50 μ m for all images. The red arrow represents the direction of shear flow.

Morphology of hydrophobic silica filled PP-minor blends after shearing



Figure S10 Optical micrographs of hydrophobic silica nanoparticle (R974) filled PP/PS blends with PS as a matrix: (a-d) PP/PS 5/95 (PP5) blends, (e-h) PP/PS 10/90 (PP10) blends and (i-l) PP/PS 20/80 (PP20) blends unfilled and filled with different concentrations (1, 3 and 5 wt.%) of R974 (R1, R3 and R5) nanoparticles after shearing at 0.1 s⁻¹ and 200 °C for 7000 s, respectively. The scale bar denotes 50 μ m for all images. The red arrow represents the direction of shear flow.

Droplet size of hydrophobic silica filled PP-minor blends after shearing



Figure S11 Dependence of the droplet size in PP/PS blends on the R974 loadings after shearing at 200 °C and 0.1 s^{-1} for 7000 s.