

## **Supplementary Information**

Figure S1. Dynamic mechanical properties of PS/SBS samples with various CR obtained by PIF processing (DMA was performed along CD with three point bending mode, as shown in inset). Glass transition temperature ( $T_g$ ) changes for (a) rubber phase and (b) PS matrix in PS/SBS samples with various CR characterized by loss modulus (E'').

To investigate the effect of the anisotropic PIF PS/SBS blends on  $T_g$ , DMA was also performed along CD with three point bending mode. Similar results of  $T_g$  of PB and PS for PIF the blends are shown as shown in Figure S1. Comparing with the non-PIF samples,  $T_g$  of PB along CD shifted to around -70°C after PIF processing Figure S1a. Meanwhile, glass transition temperature of PS phase exhibits only minor increasing after PIF processing as shown in Figure S1b.



Figure S2. Polarized fourier transform infrared spectroscopy (FTIR), spectra ranged from 600 to  $1800 \text{ cm}^{-1}$  were recorded. Polarizer was set parallel and perpendicular with respect to FD, as noted beside each curve. Pure PS samples after PIF processing (CR=2).

For pure PS, PIF processing at studied *CR* will not affect orientation of monomers, so that it's logical to attribute alignment of PS molecules to the molecule packing along FD at phase boundary, rather than stretching of molecules in bulk.