Electronic Supplementary Information (ESI)

for

Controlling Fine Touch Sensations with Polymer Tacticity and Crystallinity

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S1. AFM scan for roughness analysis. Additional larger area scans (100 μ m wide × 85 – 100 μ m) were taken on AFM to verify the roughness of all polystyrene films (Figure **S1**). As captured in smaller AFM scans of the main text, the unannealed films do not display any periodic organized structures. The larger scan reveals typical pinholes and defects for aPS and iPS films, and as mentioned in the smaller scan. Crystalline morphologies, flat-on stacked lamellae as well as some branched lamellae, are seen in the height images of annealed films. These larger area scan height



Figure S1: Larger Area Scan AFM Height images. AFM height images indicating surface topography and features across 100 μ m x 100 μ m, except for aPS which is 100 μ m × 85 μ m. Scale bar = 40 μ m.

images were used to evaluate roughness parameters, R_a and R_{pm} , the power spectrum densities, and the characteristic spacings shown in Table 1 and Figure 2a.

S2. Uniformity in crystalline direction across wafer. AFM scans were taken on annealed film, iPSA2, in the middle of the wafer and the outer edge of the wafer. The AFM height images did not show any visual differences in crystalline structures at either part of the wafer. (Figure S2) While anisotropic radial forces applied during spin-coating deposition likely did influence the crystalline growth mechanism and morphology, the resulting structures after annealing have uniform morphology and directionality at both coordinates of the wafer. Furthermore, all annealed films were first heated to a melting temperature to reduce hysteresis in chain organization caused by spinning.



Figure S2: AFM height images of iPSA2 at the middle versus edge of the wafer. Atomic force microscopy characterization of iPSA2 at two different locations along the wafer to verify location-based effects from the spin coating process. Scale Bar = $4 \mu m$

S3. GIWAXS calculation. The degree of crystallinity was calculated using the scattering profiles collected from GIWAXS measurements. All scattering profiles were normalized to a zero-intensity minimum and the degree of crystallinity was calculated by taking the area under the five prominent crystalline peaks (**Figure S3**, cross-hatched in blue) and dividing by the area of those peaks in addition to the amorphous background (**Figure S3**, cross-hatched in red), which equals the total



Figure S3: GIWAXS calculation example using iPSC2. *GIWAXS 1D scattering profiles normalized to a zero-intensity baseline for integration calculation. X-axis: scattering angle, 20. Y-axis: normalized radially-averaged intensity.*

area of coherent scattering. Area under the curves spanned from 7–23 20 and was calculated using MATLAB software.^{1–3}

S4. Surface durability. To evaluate film surface durability, microscopy images were taken for each of the films before and after human subject testing with an upright optical microscope (Nikon Eclipse LV100N POL). A bare silicon wafer (Si wafer) was also evaluated as a reference. All films appeared to remain intact after being touched and demonstrated integrity of their crystalline structures (**Figure S4**). The microscopy images for the touched samples revealed some liquid-like droplets. These small droplets are likely sebum from the subject's skin.



Figure S4: Microscopy to evaluate wear of films. Optical microscopy images of films before and after being touched in human testing. Scale bar: 10 μm.

S5. Periodicity analysis of slow wave oscillations. A fast Fourier transform (FFT) (MATLAB Software) was taken of friction traces at two representative conditions that propagated slow wave oscillations to evaluate the power of the frequency content of the friction traces. Conditions at v = 25 mm/s and M = 100g resulted in a peak at 21.05 Hz and conditions at v = 45 mm/s and M = 75g resulted in a peak at 47.01 Hz. The inverse of these frequencies provides the average periodicity of the oscillations, 0.048 s and 0.021 s, respectively. (Figure **S5**) Multiplying the periodicity of the oscillations with the associated velocity condition results in characteristic lengths of ~0.95 mm – ~1.2 mm. On average these slow wave oscillations have a correlation length of ~1 mm, or 1000 μ m. This length scale is much larger than the correlation lengths of the microstructure induced through annealing and lamellae growth, supporting that the slow wave oscillations originate from adhesion and friction mechanisms as opposed to microstructure perturbations.



Figure S5: Periodicity of friction traces. Representative friction traces at two different conditions that propagate slow frictional waves and their corresponding Fourier transforms.

References

- Marega C, Causin V, Marigo A, Chimiche S, Padova U. The Morphology, Structure and Melting Behaviour of Cold Crystallized Isotactic Polystyrene. *Macromol Res*. 2006;14(6):588-595.
- Xu H, Ince BS, Cebe P. Development of the crystallinity and rigid amorphous fraction in cold-crystallized isotactic polystyrene. *J Polym Sci Part B Polym Phys.* 2003;41(23):3026-3036. doi:10.1002/polb.10625
- 3. Schnablegger H, Singh Y. The SAXS Guide. *Ant Paar GmbH*. 2013:1-99.