Supplementary Information

Surface-Induced Alignment of Liquid Crystal Elastomers Using Rubbed Kapton® HN Films

Elaheh Asgari, *[a] Alexandre Robichaud,[b] Paul-Vahé Cicek,[c] and Andy Shih[a]
*email: Elaheh.asgari.1@ens.etsmtl.ca

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1. Morphology and Characterization of Liquid Crystal Elastomer Films

The liquid crystal elastomer (LCE) networks were synthesized using a bifunctional crosslinker (RM1: 1,4-bis[4-(6-acryloyloxyhexyloxy)benzoyloxy]-2-methylbenzene) and a monofunctional acrylate mesogen (RM2: 4-(6-acryloxyhex-1-yloxy)phenyl-4-(hexyloxy)benzoate), together with the photoinitiator 2,2-dimethoxy-2-phenylacetophenone (Irgacure® 651), in a weight ratio of RM1:RM2:initiator = 12:87:1.

Both RM2 and RM1 were obtained from Synthon Chemicals. The photoinitiator used was 2,2-Dimethoxy-2-phenylacetophenone (Irgacure® 651), sourced from Sigma-Aldrich. The prepolymer mixture was injected between two substrates coated with uniaxially rubbed Kapton® HN films, with the rubbed surfaces facing inward to promote different alignment of mesogenic units via surface-induced anchoring. All Kapton samples were prepared from 1/2 mil Kapton® HN films (Type HN, no adhesive), purchased from DuPont® Film Supplier – CS Hyde Company.

The thermal properties of this composition were verified by differential scanning calorimetry (DSC), as shown in Fig. S1, which revealed a glass transition at $T_g \approx 22.6$ °C, appearing as a smooth baseline shift characteristic of a second-order transition in LCE systems. Atomic Force Microscopy (AFM) was employed to characterize the surface morphology of three different rubbed Kapton® HN films used as alignment layers in this study. As shown in Fig. S2, each sample displays anisotropic topographical features in the form of nanoscale grooves induced by mechanical rubbing. One linear profile was extracted from each image (Fig. S2, panels a) to assess the spatial variation in surface height (Fig. S2, panels a).

As shown in Table S1, AFM areal parameters confirm distinct topographies: untreated Kapton (K-N) exhibits S_q = 3.82 nm and S_z = 56.99 nm with nearly symmetric height distribution ($S_{sk} \approx -0.05$, $S_{ku} \approx 2.70$), indicating shallow pre-

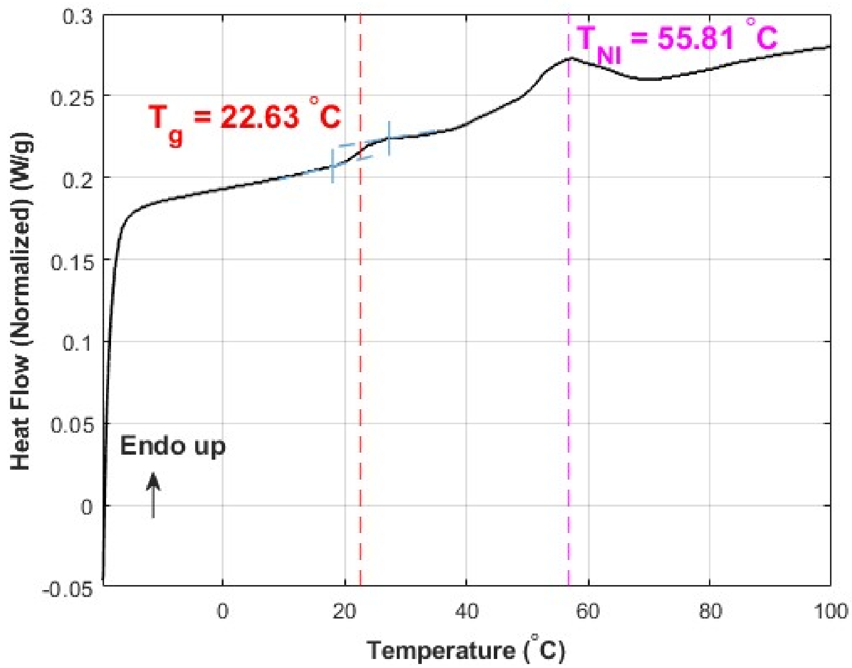

Fig. S1 DSC thermogram of a LCE synthesized from bifunctional crosslinker RM1, monofunctional acrylate monomer RM2, and photoinitiator Irgacure® 651 in a 12:87:1 weight ratio. The sample was heated from −20 °C to 100 °C at a rate of 10 °C min^{-1}. The glass transition temperature (T_g) was determined by the midpoint between the onset and completion of the baseline step, and the nematic–isotropic transition temperature (T_{NI}) corresponds to the sharp endothermic peak observed near 55 °C.

existing grooves. The sample rubbed parallel to the grooves (K-H) shows increased roughness (Sq = 8.22 nm, Sz = 76.37 nm) and a valley-biased distribution (Ssk ≈ −1.41, Sku ≈ 3.64), consistent with deeper, directional grooves. The perpendicular-rubbed sample (K-V) is the roughest, with Sq = 13.81 nm and Sz = 119.24 nm, a peak-dominated distribution (Ssk ≈ +1.54), and pronounced spikiness (Sku ≈ 6.07), reflecting the introduction of irregular asperities and intersecting scratches.

Surface area analysis further supports this trend. While the projected area (Ap) remains constant at 100 μm^2 for all samples, the developed surface area (As) increases from 100.93 μm^2 (K-N) to 101.76 μm^2 (K-H) and 102.92 μm^2 (K-V). This corresponds to the developed interfacial area ratio (Sdr), defined as the percentage increase of the actual surface area over its projected area, which rises from 0.93% to 2.92%. A higher Sdr indicates a more complex surface with greater slope content, which is expected as rubbing introduces deeper grooves and irregular peaks.

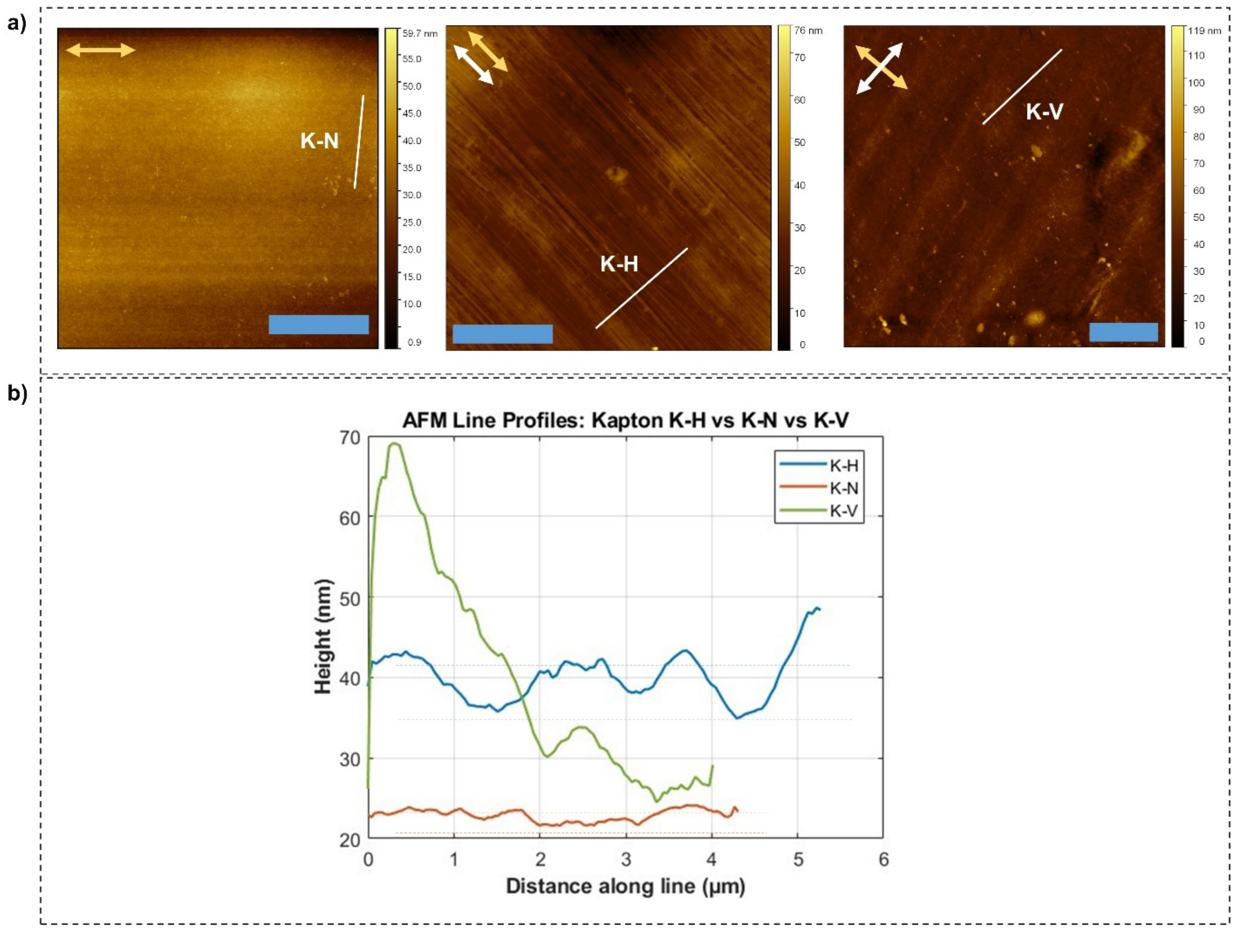

Fig. S2 a) Representative AFM line profiles for Kapton substrates: K-N, K-H, and K-V. b) Comparative plot of the extracted line profiles. K-N and K-H exhibit a sinusoidal trend (10 nm), indicating well-formed, parallel nanogrooves with uniform spacing and depth. Among them, K-H shows deeper (15 - 25 nm) and more ordered grooves. In contrast, K-V displays irregular height variations up to 40 nm along the analyzed line due to perpendicular rubbing relative to the existing grooves, resulting in cross-lines and scratches that disrupt groove uniformity.

Line profile analysis further supports these findings. K-N shows groove peak-to-valley amplitudes of approximately 10 nm, K-H exhibits periodic grooves of 15–25 nm, and K-V displays irregular height variations up to 40 nm along the analyzed line, with isolated peaks visible in the AFM image. This systematic evolution demonstrates that untreated Kapton provides a baseline with shallow grooves, parallel rubbing deepens and organizes them, and perpendicular rubbing significantly increases roughness and complexity. The differences in groove depth and uniformity, as shown in the Fig. S2, highlight the significance of rubbing direction in tailoring surface anisotropy for alignment control in LCE fabrication.

Additionally, video files showing the reversible thermal actuation behavior of the nine fabricated LCE films are provided in this documentation, with each video named according to the corresponding sample. These recordings visually demonstrate the deformation response upon heating, further supporting the alignment quality inferred from surface topography.

Table S1 Quantitative AFM areal parameters for Kapton samples: untreated (K-N), rubbed parallel to pre-existing grooves (K-H), and rubbed perpendicular (K-V). Parameters include RMS roughness (Sq), maximum peak height (Sp), maximum pit depth (Sv), total height (Sz), skewness (Ssk), kurtosis (Sku), and surface area (As) relative to projected area (Ap). These values demonstrate the systematic increase in roughness and complexity from untreated to parallel-rubbed to perpendicular-rubbed surfaces.

Parameters	**K-N**	**K-H**	**K-V**
Sq (RMS)	3.82 nm	8.22 nm	13.81 nm
Sp (max peak)	24.4608 nm	53.2612 nm	76.547 nm
Sv (max pit)	32.5278 nm	23.1132 nm	42.689 nm
Sz (max height)	56.9886 nm	76.3744 nm	119.236 nm
Skew (Ssk)	-0.0495310	-1.41438	1.53822
Kurtosis (sku)	2.70350	3.14194	6.0657
Suface area (As)	100.930 μm²	101.758 μm²	102.921 μm²
Projected area (Ap)	100.000 μm²	100.000 μm²	100.000 μm²