Dynamic Manipulation of Friction in Smart Textile Composites of Liquid-Crystal Elastomers

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Smart surfaces that reversibly change the interfacial friction coefficients in response to external stimuli enable a wide range of applications, such as grips, seals, brake pads, packaging films, and fabrics. Here, a new concept of such a smart frictional system is reported: a composite film of a plain-weave polyester textile sheet and a thermoresponsive nematic liquid crystalline elastomer (LCE). The composite is deployed with retractable microundulations of the elastomer inside each weave mesh, enabling dramatic changes of the contact interface with the opposing surface on LCE actuation, which is induced, e.g., by a change in temperature (T). At room T, the protruding viscoelastic parts of LCE in the nematic phase make contact with the opposing flat surface, resulting in a very high friction. At an elevated T (≈50 °C, isotropic phase), the undulations of LCE surface are retracted within the thickness of the textile, and the contacts are limited to small regions around overlapping textile fibers, lowering the friction dramatically. This effect is fully reversible on heating/cooling cycles. The surface undulations are spontaneous, i.e., fabricated without any lithographic or alignment techniques. The present composite opens a new way for the application of sheets/films with switchable friction enabled by stimuli-responsive LCEs.

Functional flexible polymer films and sheets[1] such as textiles, tapes, and laminating materials, are used in many applications. Their friction against other surfaces[2–4] is one of the most important factors in their practical use, since this affects the force transmission between objects, e.g., in gripping and sliding, and in tactile sensing. In many situations, the ability to switch their friction characteristics on demand[5–9] would be useful. To design such smart contact surfaces, one or more of the parameters governing the friction should be altered through reversible changes in the material components.[10,11]

The parameters controlling friction can be reviewed by considering a model for the dry friction in polymers and elastomers, which is known as the “adhesive friction model.”[2,3,12] In this model, the friction force arises from the shear strength to open the contact interface. Including the energy dissipation effect due to the viscoelastic response, and the applied sliding speed, v, the overall friction force, F, may be written as

\[ F = \alpha F_0 [1 + \phi(v)] \]  \hspace{1cm} (1)

where \( F_0 \) is the friction per unit area of real contacts at the zero crack opening, \( \phi(v) \) is a nondimensional factor expressing the velocity-dependent viscoelastic dissipation, and \( \alpha \) is the ratio of the real contact area to the apparent one (0 < \( \alpha < 1 \)). \( F_0 \) is related to the static interfacial energy of the two materials in contact. The geometric factor \( \alpha \) should depend on the applied apparent normal pressure, \( P \) (or load \( L \)), and usually shows a monotonic increase with \( P \), known as the “Amontons law.” Additionally, the variation in the elastic modulus of the materials in contact, which also affects the interfacial deformation, may be incorporated in \( \alpha \). For the case of friction on the rough surface, the local topography effect can also be included in \( \alpha \); the smaller or larger value should be assumed for the corrugated or smooth surface, respectively.

According to Equation (1), there are, in principle, multiple parameters available to tune the friction. If materials in contact are changed, \( F_0 \) may change via the altered surface energies; parameters \( \alpha \) and \( \phi(v) \) also may change via the altered storage and loss moduli, respectively. If the surface topography is modulated, \( \alpha \) is directly affected as well. Therefore, surface designs that induce drastic changes of materials in contact and the surface topography, will provide friction with a wide dynamic range.

For this purpose, ideas mainly relying on the surface topography changes have been explored, using mechanical buckling-based surface wrinkles[7–9,13] and fingerprint textures of a chiral nematic polymer coating.[5,6,14] The former system requires the application of macroscopic strain to the sample to generate the microscopic surface wrinkling, which makes it not useful as freestanding sheets. Although the latter liquid crystal polymer systems[15,16] are attractive, the good wrinkling effect requires...
specific alignment layers for the polymer on the surface, which is technically challenging and only useful in limited settings. Liquid crystal elastomers (LCEs) are very promising systems to induce very large topographical changes via phase transitions\(^\text{[17–25]}\) that can be triggered by multiple stimuli, such as temperature, light, and chemicals. In addition, the LCEs show drastic changes in the bulk viscoelasticity\(^\text{[18,26–30]}\) which allows to tune friction via the \(F_0\), \(\alpha\), and \(\phi\) entries in the basic Equation (1). However, the development of the dynamic frictional system through the combination of these effects remains an open challenge.

Here, we propose a new composite system, combining a polyester textile and thermally responsive nematic LCEs. The generic idea is to simultaneously change the contact area through the topographic effects that influence the magnitude of \(\alpha\), change materials in direct contact, which affects \(F_0\) and \(\phi\), and also use the dynamic soft-elasticity of a nematic LCE phase to separately affect \(\phi\) via the enhanced internal dissipation. All of these changes occur on LCE actuation upon an external stimulus, e.g., temperature change that induces the phase transition of incorporated LCEs. Then, Equation (1) is rewritten indicating the relevant variables as

\[ F(T, L, v) = \alpha(T, L, v) F_0(T) \left[1 + \phi(T, v)\right] \]  

(2)

The proposed changes become possible through a fine geometrical design of thermally retractable microundictions of LCE “pockets” formed in each opening of the textile mesh. They are also designed to be fabricated through a simple lithography-free deposition method. At room temperature (RT), the buckled viscoelastic LCE parts in the nematic phase protrude out of the composite film and make contact with the opposing surface over a large area, resulting in high friction. In contrast, at high temperature (HT) that turns the LCE isotropic, the undulations of the elastomer are flattened and retracted within the confines of the textile mesh. As a result, the contacts with the opposing surface are limited to small regions around overlapping points of the hard textile fibers, lowering friction dramatically.

Our system is the composite (Figure 1) of a nematic LCE\(^\text{[23,25,30]}\) (Figure S1, Supporting Information) and a plain-weave textile, in which the volume other than the fiber components within the thickness of the textile is almost fully filled with the LCE (Figure 1b), as in the rubber-coated textiles. The basic properties of the present LCE have been characterized using the neat LCE samples, and listed in the Supporting Information: polymerization reactions (Figure S2, Supporting Information), nematic-isotropic transition \(T_{NI}\) (Figure S3, Supporting Information), LC phase (Figures S3 and S4, Supporting Information), thermal actuation (Figure S5, Supporting Information), and viscoelastic response (Figure S6, Supporting Information). In brief, the LCE has \(T_{NI} \approx 35 ^\circ\text{C}\) and the glass transition temperature of \(\approx -40 ^\circ\text{C}\), spontaneous and reversible changes of the shape with the uniaxial strain of at least 30%, and of the soft viscoelasticity with the high loss factor tan\(\delta\) in the nematic phase.\(^\text{[30]}\)

The fabrication of the composite begins with molding the LCE precursor and the textile sheet between two flat glass slides under normal pressure,\(^\text{[31]}\) and its thermal curing in the isotropic phase (Figure 1a, Methods). The first thiol-ene reaction, creates a random isotropic polymer network. During annealing of the peeled freestanding sample, the solvent is evaporated, causing the volumetric shrinkage. Since the textile lattice is locally rigid, the shrinkage occurs exclusively in the direction

![Figure 1](https://example.com/figure1.png)

**Figure 1.** Preparation of the LCE–textile composite sheet with microshape changeability. a) The LCE precursor with the textile between two glass slides undergoes the first cross-linking reaction at elevated temperature (isotropic phase). b) Drying the peeled sample under vacuum, prompting solvent evaporation and volume contraction. c) The second network crosslinking by UV irradiation under local uniaxial strain at RT (nematic phase). The LCE parts buckle protruding in one of the out-of-plane directions, “up” or “down,” alternatively along the nearest neighboring mesh openings. This structure is “memorized” by the secondary crosslinking in the nematic phase. d) Release of applied tensile strain and annealing in isotropic phase causes restoring of the original shape, memorized in the isotropic state of LCE and the original textile shape with the square mesh opening. After cooling back to RT, the further modulated undulations of LCE segments appear due to the mismatch between the square shape of the mesh and the secondary memorized shape.
normal to the sample plane (Figure 1b), which is different from the neat LCE systems without constraints of the textile frame.

Then, the sample is uniaxially stretched at RT (in the nematic state) to the strain of 30% in the diagonal direction of the square mesh openings (Figure S7, Supporting Information), which is the most compliant direction (x) of the plain-weave textile. Under this strain, the LCE parts at mesh openings adopt a buckled surface structure in response to the induced local compressive strain (Figure S8, Supporting Information) of \( \approx -45\% \) in the orthogonal direction (y) (Figure 1c; Figure S9a, Supporting Information).

Two energetically equal states of buckling appear: bending “up” or “down,” which is confirmed by the observation of both sides at the same location (Figure S9b,c, Supporting Information). In most parts on the sample, the buckling directions show a regular square pattern, with the nearest neighboring mesh openings shown one opposite to each other. Since the LCE is continuous across neighboring meshes, the alternation of the buckling direction may minimize the bending energy of LCE parts between the mesh openings. In addition to this structural change, the complicated alignment of the nematic director in this buckled state should emerge. This state of the nematic director alignment coupled to the shape is then memorized by the UV-induced second polymerization process that stabilizes the nematic phase.

After the second photo-crosslinking, the applied strain is released and the sample is annealed to equilibrate in a load-free state (Figure 1d). At HT, the original memorized state of the isotropic LCE is restored, and the original textile shape with square mesh opening is also recovered. After cooling back to RT, the modulated undulations of LCE segments, reflecting those formed under the applied strain, appear without changing the square shapes of the mesh. Owing to the mismatch between the naturally square shape of the mesh openings, and the rhombic shape recorded in the nematic phase, additional buckling of the elastomer surface occurs in response to the emerging compressive strain in the x direction. The results also indicate that the additional buckling has lower energy than the simple elongation of LCE parts together with mesh shape deformation without undulation of LCE parts. Since the initial undulations can assist in nucleating the additional buckling, the initial buckling directions memorized under applied strain are retained in the structure (Figure S9a,b, Supporting Information). Upon heating again to \( T > T_{NI} \), the undulations retract, restoring the state of the first crosslinked network. Consequently, the temperature change mainly induces reversible alternation of microundulations, while the macroscopic shape of the composite sheet remains approximately constant (Figure S10, Supporting Information).

Since buckling of the LCE segments is the key phenomenon behind the microundulations, the effective aspect ratio, (lateral length)/(thickness), of the LCE segments in the mesh openings is an important parameter (Figure S11, Supporting Information), which depends on the mesh size. In the subsequent study, the composites of the textile with the mesh opening of 0.199 mm (T100) are used as the representative system with ideal microundulations.

The detailed temperature-dependent surface topographies are shown in Figure 2 (with details in Figures S12 and S13 in the Supporting Information). The undulations of the LCE parts in the mesh openings at RT (nematic) are retracted at HT (isotropic) (Figure S14, Supporting Information) toward the mid-plane of the sample sheet, and flattened. This is also traced by the diffusion of laser light transmitted through the sample\(^{38} \) showing the broadening of the diffusion peaks due to the roughening of the interface at RT (Figure S15, Supporting Information).
The amount of the solvent during the preparation, \( c_{\text{sol}} \), affects the surface topography via the final thickness of the LCE layer. In particular, it controls the height difference \( h \) between the fiber-crossing points and the LCE parts (Figure 2g,h). At HT, \( h \) linearly increases at \( \approx 0.5 \, \mu m \, wt\%^{-1} \) as shown by the red dashed line in Figure 2h. Since the textile half thickness is \( \approx 5 \, \mu m \), the result indicates that the shrinkage of LCEs near the center of the mesh opening occurs mainly in the direction perpendicular to the film. The nonzero value of \( h \) at \( c_{\text{sol}} = 0 \) wt\% would be due to the non-negligible curing shrinkage (syneresis)\(^{[33]}\) of \( \approx 10\% \) during the present polymerization. At RT, values of \( h \) become smaller due to the microundulations of the LCEs. The transformation between two states was fully repeatable over at least ten cycles (Figure S13 and Movie S1, Supporting Information), as expected from the neat sample (Figure S5, Supporting Information) and the other nematic LCEs.\(^{[17–25]}\)

The static, \( F_S \), and kinetic friction forces, \( F_K \), between the LCE–textile composite and the flat glass surface were investigated (Figure 3a) at the range of load \( L < 1 \, N \), for which the deformation of the polyester fiber at the overlapping point is negligible (Figure S16, Supporting Information). The typical time-dependent frictional signals are shown in Figure 3b, in which \( F_S \) and \( F_K \) are defined. The temperature-dependent static and kinetic friction coefficients, \( \mu_S \) and \( \mu_K \) (Figure 3c,d), are obtained as slopes of the linearly fitted line to the load-dependent friction force. They are roughly classified as either the higher \( (\mu_S > 1, \mu_K > 0.6) \) or lower ranges \( (\mu_S < 0.4, \mu_K < 0.2) \). According to the observation of contact regions (Figure S18, Supporting Information), the high friction corresponds to the states with the larger contact area consisting of LCE parts (leading to a larger \( \alpha \) (Figure S18b–d, Supporting Information). In the low friction case, contacts only occur at points of overlapping fibers, with small area (Figure S18e–g, Supporting Information). In particular, the sample prepared with 10 wt\% solvent shows maximum T-dependent changes in both \( \mu_S \) and \( \mu_K \), each of which shows almost sixfold increase on cooling from HT to RT. For comparison, the best results on the increase of friction in a LC polymer layer were at most twofold\(^{[6,14]}\) and only when two patterned surfaces were against each other. In the deformation-induced wrinkled bilayer surface\(^{[7,8]}\) the friction increase was at most 1.5-fold, see the review\(^{[13]}\) for detail. The results demonstrate the effectiveness of the present composite design with T-dependent topographical changes on the dynamic friction.

Now let us briefly discuss the observed difference between \( F_S \) and \( F_K \) illustrated in Figure 4, as functions of the normal load, and the sliding speed. In general, \( F_S \) at \( L = 0 \) is of the same order of magnitude as the adhesive strength, \( \sigma_{\text{ad}} \). Since \( \sigma_{\text{ad}} \) between the glass surface and the LCE at RT after the dwell time of \( \approx 30 \, s \) is in the order of 0.1 MPa,\(^{[30]}\) \( F_S \) at \( L = 0 \) with the apparent area of interface \( (A = 133 \, mm^2) \) can be estimated as \( \alpha A \sigma_{\text{ad}} = 1 \, N \), where \( \alpha = 0.1 \) is the typical ratio of the contact area at \( L = 0 \) to \( A \). This corresponds roughly to the nonzero values of the fitted line on the experiments at RT on 0 and 10 wt\% solvent systems at \( L = 0 \), giving \( \approx 0.9 \, N \) (Figure S17a, Supporting Information) and \( \approx 0.4 \, N \) (Figure 4a), respectively. The effect becomes smaller for the kinetic frictions (Figure 4b), which show lines through the origin at \( L = 0 \), because of the interfacial sliding; no dwell time for the contact area growth.

Both \( F_S \) and \( F_K \) also increase with the sliding velocity \( v \) (Figure 4c,d) while at constant load \( L \). The difference is most

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**Figure 3.** Friction tests. a) Schematic for friction tests. b) Example of the friction signal and the definition of static and kinetic frictions, \( F_S \) and \( F_K \), respectively. The data on the sample prepared with 10 wt\% solvent at \( v = 5 \, mm \, s^{-1} \) with \( L = 0.3 \, N \) (apparent normal pressure of 2.2 kPa) are shown. c) Static, \( \mu_S \), and d) kinetic, \( \mu_K \), friction coefficients, obtained as slopes of plots of friction force versus normal load.
striking at RT. The majority of the frictional interface is assumed to be the viscoelastic nematic LCE surface, with tan δ of ≈0.7 or above (Figure S6, Supporting Information), which should boost viscous dissipation, φ, in Equation (2). In contrast, in the isotropic phase at HT, the contact area is greatly reduced. Moreover, the LCE in contact is in the isotropic phase with the lower tan δ ≈ 0.3 or below, and the hard polyester surface at the fiber-crossing points is in contact, which supports most of the load. Note that at RT the friction of the sample prepared with 10 wt% solvent is only slightly larger than that observed on the neat textile (Figure 3c,d). Thus the slight difference would originate from the modest contact with the isotropic LCE parts around the fiber-crossing points (Figure S18e, Supporting Information). Overall, the present dynamic friction is qualitatively explained by T-dependent changes in α and φ in Equation (2), which are related to the contact area and viscoelastic property, respectively.

In general, the friction depends on the properties of two surfaces in contact. The present temperature-induced drastic change in friction may be limited to the cases, in which the hard materials with a flat surface are sliding. If, for example, soft materials with surface roughness comparable to the present mesh periodicity are used as the counter surface, the larger contact may be possible even at HT on the sample prepared with 10 wt% solvent, and thus, resulting in the reduction of the dynamic range of the friction. The easy way to optimize the system is to increase the value of $h$ (Figure 2h), according to the deformability and roughness of the counter surface. In addition, the size of the mesh opening and the fiber diameter (Figure S11, Supporting Information), which also should affect $h$, can be tuned for this purpose. Fatal wear should also be minimized. First, the fundamental wear resistance of the material components, especially, of viscoelastic soft LCEs should be improved. Additionally, the good adhesion between LCEs and textile fibers may be required. Future work toward practical applications would focus on these issues with the help of the matured technologies known to ordinary rubbers and their composites with textiles and fibers.\(^{[34]}\)

In summary, here we report the design of LCE–textile composites with thermally retractable microundulations for the dynamic manipulation of friction coefficients via modulation of the contact states. The buckling-based spontaneous undulations of LCE parts in the openings of the textile mesh develop through a simple lithography-free procedure. The T-dependent interfacial contact states of the LCE–textile composites to a flat glass surface can be tuned by controlling the fine structure. With the optimized design, the LCE–textile composite makes contact at undulated viscoelastic LCE parts in the nematic phase at RT. In contrast, at HT, textile fibers at their crossing points mainly constitute the contacts with the small area. This alternation, which includes the changes in the contact area and replacement of the materials in contact, can modulate friction forces by sixfold. Owing to the simple fabrication of the transformable microstructure, and the possibility of the further optimization, including the fine structural modifications and tuning of LCE properties, the present design could provide a route to smart LCE–textile composite sheets and films that can reversibly change friction on demand. Since the textile\(^{[34]}\) provides the LCE with a better mechanical strength, while retaining the flexibility of a freestanding film, the present composite would also be promising for handling and further processing in a wide range of applications, e.g., robot hands, touch screen devises, and smart textiles.

**Experimental Section**

**Materials and Preparation of LCE–Textile Composite:** For the preparation of LCE, the methods reported previously were followed,\(^{[30,31,34]}\) with two steps of crosslinking reactions: a thiol-acrylate Michael addition and a photoinduced radical polymerization of diacrylates, with slight modifications. The diacrylate monomer, 1,4-bis-(4-(acyloyloxyhexyloxy)benzoyloxy)-2-methylbenzene (RM82), was purchased from Wilshire Technologies (Figure S1, Supporting Information). The diacrylate spacer, tri(propylene glycol) diacylate (TPGDA), and two thiol monomers: 2,2′-(ethyleneoxy)diethanethiol (EDDET) and pentaneethanol tetras (3-mercaptopropionate) (PETMP), were purchased from Sigma Aldrich. Triethylamine (TEA, Sigma Aldrich) was used as the catalyst of the Michael-addition thiol-ene reaction. Irgacure2959 from BASF was used as the photoinitiator for the radial polymerization of acrylates. As the radical scavenger, butylated hydroxytoluene (BHT, from Sigma Aldrich) was used to suppress the unwanted reaction before UV light irradiation. Toluene (Sigma Aldrich) was used as the solvent. All chemicals were used in their as-received condition with no purification. Three plain-weave polyester (polyethylene terephthalate) textiles composed of a fiber with the diameter of 0.055 mm and different mesh openings (0.368 mm (T60), 0.199 mm (T100), and 0.086 mm (T180)) were purchased from Yamani inc. Japan.

At the specific molar ratio of functional groups shown in Figure S1 (Supporting Information), RM82, TPGDA, EDDET and PETMP were weighted, Irgacure2959 was added at 0.2 wt%, BHT was added at 0.5 wt%, and toluene was added at 0, 10, and 20 wt%. By increasing the amount of TPGDA, it was possible to lower $T_{NI}$ and here it was adjusted to make $T_{NI} = 35 ^\circ C$. Note that the amount of the solvent was important to control the final thickness of LCE at the mesh openings, and thus varied here to study the effect. After the mixture was gently mixed at an
elevated $T = 70\,^\circ\mathrm{C}$ for $\approx 10\,\text{min}$, TEA was added at 1.5 wt% to start the Michael-addition reaction between thiol and acrylate groups. The mixture was kept between two glass slides with a textile mesh with thickness of $\approx 0.1\,\text{mm}$ at $70\,^\circ\text{C}$ (isotropic phase) overnight under a pressure of $\approx 10\,\text{kPa}$ applied to the direction normal to the glass surface to squeezing out the excess liquid. The sample was released from the glass mold, and placed at $80\,^\circ\text{C}$ in vacuum to remove the solvent. The sample was then cooled down to RT and UV light (365 nm) was applied for 20 min under a uniaxial tensile strain to finalize the crosslinking between the rest of the acrylates. The tensile strain of 30% was applied in the direction of 45° from the yarn direction; each mesh of the textile was thus elongated to a rhombic shape. The locally deformed and aligned state of the LCE segments were memorized at this crosslinking step. Upon releasing the strain, microundulations of the LCE segments were finally formed. For WAXS and mechanical measurements, samples without the mesh were also prepared, and UV light was applied under uniaxial tensile strain of 50% as reference. The samples were annealed at 80 °C in vacuum oven for 2.2 kPa) was gently out the excess liquid. The sample was released from the glass mold, and placed at 80 °C oven for 24 h after addition of TEA was also prepared. After taking the spectrum, UV light was irradiated for 20 min for evaluation of the final state. The conversion ratios at the end of each reaction were defined as $\xi = \frac{\sigma_{\text{final}} - \sigma_{\text{initial}}}{\sigma_{\text{initial}}}$, where $\sigma_{\text{initial}}$ and $\sigma_{\text{final}}$ are the initial and final peak area of each functional group, thiol, or acrylate.

### Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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### Conflict of Interest

The authors declare no conflict of interest.

### Keywords

buckling, friction, interfacial contacts, liquid crystal elastomer, textile mesh

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