

Research Article

Thermoresponsive biomimetic earthworm polymeric mucus for smart lubrication

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Abstract: Oil-based lubrication is widely used in industrial robots and specialized unmanned aerial vehicles (UAVs), yet frictional heating and elevated-temperature operation often accelerate lubricant depletion and lubrication failure; converting such adverse thermal stimuli into adaptive self-lubrication remains a key challenge. Inspired by the friction-stimulated, mucus-mediated self-lubrication of earthworm skin, a dynamic covalently cross-linked oleogel was fabricated by integrating heterogeneous gradient-wettability materials with a metastable lubricant material

matching effect. The oleogel shows robust interfacial adhesion (8.56 MPa) and toughness (8.7 MJ m⁻³) and operates as a thermoresponsive wettability switch that releases an earthworm-like polymeric “mucus” lubricant. Temperature-induced sol-gel transition of the network, combined with capillary adsorption in surface and internal micro-nano pores, enables more than ten reversible cycles of lubricant locking, exudation, and recovery, thereby establishing an intelligent self-lubrication mode based on an effective conversion from a hydrophilic to a hydrophobic interfacial state. Under this thermosensitive switching, the oleogel adaptively transitions between solid-solid and solid-liquid contact; at room temperature the low-viscosity state affords a moderate coefficient of friction ($\mu = 0.123$), whereas thermal activation yields a low-friction self-lubricating state ($\mu = 0.055$). This work provides theoretical guidance and technical support for smart lubrication of robotic joints and for polymer designs that accommodate multiple friction regimes.

Keywords: Bioinspired earthworm; Polymeric mucus; Thermo-responsive stimulus; Oleogel; Smart lubrication

1 Introduction

Industrial robots and specialized unmanned aerial vehicles rely on rotating components that must deliver adaptive and controllable tribological performance under complex and harsh service conditions, which remain a major challenge [1]. Mechanical friction can generate localized heat, thereby increasing the coefficient of friction (μ) and ultimately shortening component lifetime. Oil-based lubrication is the conventional solution; however, under operating conditions exceeding 120 °C, lubricants at interfaces such as joint bearings or universal couplings are prone to severe oxidation, volatilization, and viscosity loss. These changes hinder the formation of a continuous lubricating film and accelerate wear [2, 3]. Therefore, under the coupled effects of frictional self-heating and elevated ambient temperature, developing intelligent lubricating materials that can sense

service conditions and actively regulate interfacial friction by leveraging environmental temperature represents a promising strategy for temperature-sensitive applications. The development of smart interfacial materials has been inspired by stimulus-responsive behaviors observed in plants and animals in nature, motivating the design of surface-functional materials that respond to stimuli such as temperature, shear rate, pH, humidity, ionic strength, and related environmental cues [4].

In nature, many organisms have evolved unique environment-responsive surfaces [5, 6] that can reversibly regulate interfacial contact modes (frictional or lubricated) to mitigate risks. For example, earthworms and slugs [7, 8] encounter excessive resistance when moving through soil. To prevent damage to the fragile skin tissue at the solid–solid interface ($\gamma_{\text{skin-soil}}$), earthworms secrete copious mucus from subcutaneous glands. This mucus, composed primarily of water, carbohydrates, proteins, lipids, and polysaccharides [9], transforms the contact mode from a solid-solid ($\gamma_{\text{skin-soil}}$) to a solid-liquid ($\gamma_{\text{mucus-soil}}$) interface. The mucus layer not only protects the skin surface but also imparts adaptive lubrication, effectively reducing friction [10]. Although the specific triggers vary (e.g., mechanical stimuli, humidity, or other environmental signals), the underlying principle is essentially the same: external stimuli regulate the formation and/or replenishment of a lubricating layer, thereby converting a frictional interface into a lubricated one. This bioinspired concept of “stimulus-regulated switching” has been translated into synthetic gels actuated by different types of stimuli. Inspired by the muscle-stiffening-triggered lubrication of long-snouted catfish, Zhou et al. [11] developed a bilayer hydrogel polymer with modulus-adaptive properties, in which thermally induced phase separation enables a self-lubrication model that switches between high- and low-friction states. Similarly, Cui et al., inspired by the self-lubricating layer of earthworm skin, designed an “breath pattern” structure in which discrete lubricant reservoirs are embedded in a polymer matrix [12]. Mechanical stimuli induce lubricant release, thereby lowering friction. These examples highlight that mimicking the stimulus-responsive behaviors of biological skin to construct adaptive

gels capable of switching between frictional and lubricated states has emerged as a current research focus.

Dynamic covalent chemistry [13-15] and supramolecular interactions [16] are versatile strategies for imparting materials with self-healing capabilities [17-19], stimulus responsiveness [20-22], and tunable mechanical properties [23-25]. The kinetic equilibria of dynamic covalent bonds can be precisely modulated by external stimuli such as light [26-28], mechanical force [29-31], temperature [32-36], and solvents [37-39]. In addition, weak intermolecular interactions, including hydrogen bonding [40-43], metal-ligand coordination [44-47], π - π stacking [48-50], host-guest recognition [51-54], and electrostatic interaction [55], can be strategically combined to enable the controllable construction of gels with multiple self-lubrication modes. The design of such multi-mode friction/lubrication, stimulus-responsive gels is strongly influenced by the surrounding medium, which may include water, oils, ionic liquids, and organic solvents. Zhao, Liu, and co-workers [56] reported a temperature-responsive, phase-separating ionic gel that achieves triple frictional states through structural evolution of the gel matrix and the distinct adsorption kinetics of the ionic liquid. In contrast, developing responsive organogels (oleogels) with excellent matrix compatibility remains challenging because of the combined effects of polarity mismatch, surface tension disparities, and high viscosity at the oleogel interface. Zhou et al. [57] addressed this challenge by tailoring the compatibility between a dynamic covalent network derived from the ring-opening polymerization of thioctic acid (TA) and lubricating oil, thereby creating an adaptive oleogel capable of reversible gel-sol transitions. Liu and Wang et al. [58] achieved macroscopic superlubricity by compounding porous polyimide with an oleogel incorporating modified molybdenum disulfide (MoS_2), where the synergistic action of four lubrication and friction-reduction mechanisms yielded a coefficient of friction as low as 0.007. Nevertheless, realizing hydrophilic-hydrophobic wettability switching in intelligent, responsive oleogels remains challenging because the nonpolar oil phase screens/solvates many weak interactions and reduces stimulus sensitivity,

while wettability switching requires interfacial compositional rearrangement that is kinetically hindered by a viscous, crosslinked matrix. Moreover, introducing polar motifs to enable hydrophilicity often compromises oil–matrix compatibility, increasing the risk of phase separation, uncontrolled oil bleeding, and loss of mechanical robustness, especially at elevated temperatures.

Here, inspired by the mucus-releasing behavior of the soft skin of earthworms during in-soil locomotion under frictional stimulation from the surrounding environment (Fig. 1a), a series of gradient-wettability material combinations were rationally designed (Fig. 1b). The design incorporates (1) polyethylene glycol (PEG) to provide hydrophilicity; (2) polytetramethylene ether glycol (PTMEG) to provide hydrophilicity and moderate modulus; (3) hydroxyl-terminated polydimethylsiloxane (HO-PDMS-OH) to offer low polarity compatible with the lubricant medium; and (4) silicone oil as the lubricant. Through the use of the dynamic crosslinker diaminoethanedioxime (DAG), these hydrophilic–hydrophobic heterogeneous structures were effectively integrated and optimally matched with the lubricant medium, enabling the controlled fabrication of dynamic covalently cross-linked oleogels (DCP-x vol.%, $x = 10, 30, 50$). In the dynamic covalent network, the low-polarity phase stores the critical volume fraction of lubricant in a metastable state within localized regions of the gel. Upon temperature stimulation, the kinetic dissociation of the dynamic oxime–urethane bonds induces a sol-gel transition, and disruption of the metastable system weakens the crosslinked network, triggering lubricant release (Fig. 1c). Owing to this thermo-responsive effect, the oleogel surface exhibits reversible hydrophilic, adhesion-governed behavior and hydrophobic, slip-governed behavior under cyclic high-low temperature conditions, enabling adaptive switching between solid-solid contact and solid-liquid contact modes, with the coefficient of friction decreasing from a moderate value of 0.123 to a low value of 0.055 (Fig. 1d). Notably, this design concept is inspired by the earthworm’s natural behavior: under normal locomotion, the skin exhibits a moderate μ , defined as the hydrophilic, weak-adhesion state. When stimulated by adverse environmental factors, the earthworm secretes mucus on its skin, enabling a

transition from solid-solid (γ_{s-s}) to solid-liquid (γ_{s-l}) contact, thereby greatly reducing the surface μ —defined as the hydrophobic slip state. By combining gradient-wettability materials, appropriately matched lubricants, and a dynamic covalent crosslinking network, this strategy achieves bioinspired, controllable switching between hydrophilic and hydrophobic interfacial states, yielding an intelligent, self-lubricating behavior in the oleogel system (Fig. 1e).

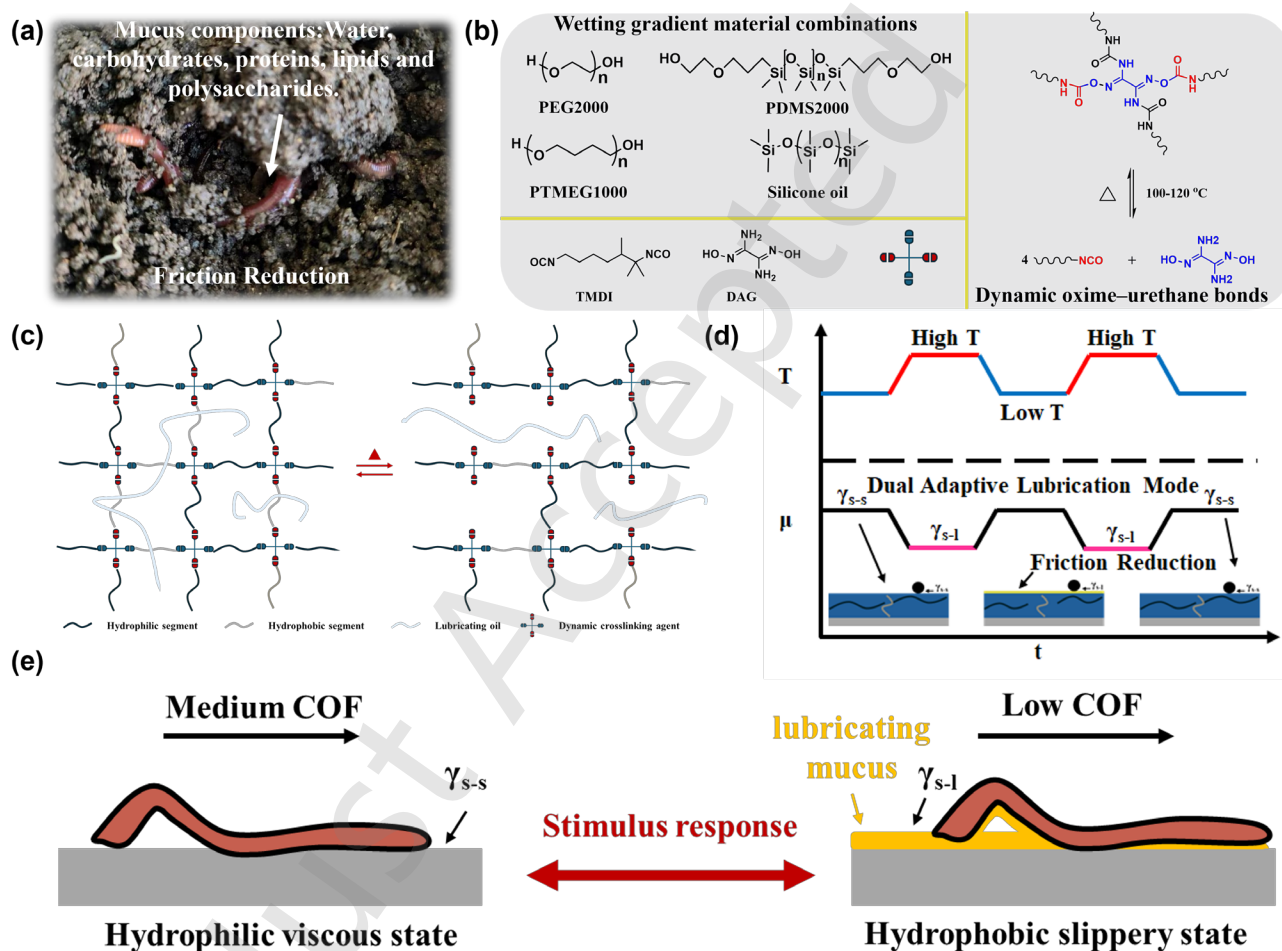


Fig. 1 Schematic of an earthworm-inspired, dynamically cross-linked intelligent self-lubricating oleogel that enables controllable switching between hydrophilic and hydrophobic interfacial states. (a) Constituents of earthworm mucus. (b) Controlled construction using gradient-wettability components in synergy with dynamic covalent cross-linkers. (c) Thermally induced exudation of the lubricant medium from the dynamic covalent network. (d) Temperature-responsive intelligent lubrication modes. (e) Stimulus-responsive transitions between lubrication modes in the earthworm-inspired system.

2 Experimental section

2.1 Materials

Polytetramethylene Ether Glycol ($M_w = 1000 \text{ g mol}^{-1}$), Polyethylene Glycol ($M_w = 2000 \text{ g mol}^{-1}$), Carbinol(hydroxyl)terminated polydimethylsiloxanes ($M_w=2000 \text{ g mol}^{-1}$) and dibutyltin dilaurate (DBTDL, 95%) were purchased from Macklin, China. Tetrahydrofuran (THF, Extra Dry, Water $\leq$ 30ppm) was purchased from Energy Reagent, China. Diaminoglyoxime (95%), *N-N*-Dimethylacetamide (DMAC) and Trimethylhexamethylene Diisocyanate (TMDI, 97%) were obtained from Aladdin Reagent, China. The silicone oil was purchased from Tianjin Zhonglian Chemical Reagent Co., Ltd. Ethanol and THF were purchased from XILONGS Scientific, China. Hydraulic oil and machine oil were obtained from commercial sources. All chemicals were used as received.

2.2 Synthesis of DCP

PTMEG (1 g, 1.00 mmol), PEG (1 g, 0.50 mmol), and HO-PDMS-OH (1 g, 0.50 mmol) were heated under vacuum at 120 °C for 1 h to remove residual moisture. After cooling to 50 °C, TMDI (1.26 g, 6.00 mmol) dissolved in 20 mL of anhydrous THF was added dropwise via a constant-pressure dropping funnel, and the reaction was allowed to proceed for 12 h to yield the prepolymer. Subsequently, DAG (0.47 g, 4 mmol) dissolved in 10 mL of DMAC was introduced into the prepolymer solution in a three-neck flask via syringe. The reaction was conducted at 60 °C for 12 h to obtain the dynamically crosslinked polymer (DCP). THF and DMAC were removed using a rotary evaporator, and the transparent, viscous polymer melt was redissolved in tetrahydrofuran. DCP was fabricated into a coating by dip coating and subsequently cured at 70 °C for 24 h.

2.3 Synthesis of DCP-x vol.%

At the initial stage, silicone oil was added at varying volume fractions (10%, 20%, 30%, 40%, 50%), with subsequent steps following the same procedure as for the preparation of the DCP samples.

2.4 Characterization

Proton nuclear magnetic resonance (^1H NMR) spectra were recorded on an AVANCE III HD 400 MHz spectrometer using DMSO- d_6 as the solvent and tetramethylsilane (TMS) as the internal reference. Fourier transform infrared (FTIR) spectroscopy was performed using a Bruker Invenio spectrometer over a wavenumber range of 4000–400 cm^{-1} with samples prepared as KBr pellets. Experimental data were subjected to baseline correction and normalization. Sample transparency was evaluated using a UV-6300 double-beam spectrophotometer. The water contact angle (WCA) was measured using a DSA-XROLL instrument equipped with a 5 μL water droplet. Samples treated at 100 $^\circ\text{C}$ were allowed to equilibrate at room temperature for 5 min prior to measurement. Differential scanning calorimetry (DSC) was conducted using a Shimadzu DSC-60A Plus to investigate the thermal properties of the samples. Measurements were performed from -70 to 150 $^\circ\text{C}$ at a heating rate of 10 $^\circ\text{C min}^{-1}$ under a nitrogen atmosphere (flow rate: 50 mL min^{-1}). Thermogravimetric analysis (TGA) was performed using a Shimadzu TGA-51H to assess the thermal stability of the samples. The instrument was purged with nitrogen at a flow rate of 50 mL min^{-1} , and samples were heated from ambient temperature to 600 $^\circ\text{C}$ at a rate of 10 $^\circ\text{C min}^{-1}$. The crystallinity of the samples was investigated using an X-ray diffractometer (Bruker D8 Advance) with $\text{Cu K}\alpha$ radiation, scanning over a 2θ range of 5 – 60° . The oil storage, release, and recovery processes of the oleogel, as well as its surface morphology before and after self-healing, were observed using an MV3000 optical microscope. The microstructure of dynamically crosslinked polymers and oleogels was examined using a desktop scanning electron microscope (Phenom XL).

2.5 Mechanical testing

Uniaxial tensile and cyclic tensile tests were conducted using a universal testing machine (WL2100C, Withlab, South Korea). Dumbbell-shaped specimens (12 mm length, 2 mm width, ~ 0.8 mm thickness) were prepared, and tests were performed at a strain rate of 50 mm min^{-1} .

2.6 Rheological testing

Rheological measurements were carried out using a TA DHR-2 rheometer. Samples with a thickness of 1 mm were tested under 5% strain at various temperatures. On the other hand, the temperature-dependent viscosity of the silicone oil was measured using a touchscreen digital viscometer (LC NDJ99T, LICHEN, China).

2.7 Self-healing and adhesion test

Self-healing tests were conducted by manually scratching the surface with a knife, followed by thermal treatment to repair the scratched area, with heat-assisted self-healing performed at 100 °C. Pull-off tests were performed using an automated tester (Elcometer 506 Digital Adhesion Tester) in accordance with ASTM D4541-09. All samples were coated on copper, steel, and glass substrates for adhesion testing. The pull-off rate was set to 0.2 MPa s⁻¹. The average adhesion strength was calculated based on measurements from three distinct regions of the coating.

2.8 Tribological testing

An MS-T3001 tribometer (Lanzhou Huahui Instrument & Equipment Co., Ltd., China) was used to evaluate the tribological performance of the oleogels. A ball-on-disk configuration was adopted with an alumina ball of 2 mm diameter sliding against a disk of $\Phi 45 \times 8$ mm. Tests were conducted under unidirectional rotation with a friction radius of 3 mm and a constant normal load of 0.2, 0.5, 1, 2 N. The Stribeck curves were constructed by plotting the friction coefficient as a function of $\log_{10}(\eta U / p)$. The sliding speed was calculated from the track radius $r = 3$ mm as $U = 2\pi r \cdot rpm / 60$. The calculated apparent contact pressures were approximately 18.1, 24.4, 30.7, and 38.6 MPa under normal loads of 0.2, 0.5, 1, and 2 N, respectively. Temperature-dependent friction tests were performed using two protocols. For the intermittent temperature-switching test in Fig. 3g, the oleogel-coated sample was first tested at 25 °C. The test was then paused, and the sample was heated on a heating stage to 85 °C, followed by continued friction testing under the same conditions. Subsequently, the test was paused again, and the sample was naturally cooled to 25 °C before the next friction segment. This stepwise 25–85–25 °C cycle was repeated to evaluate the reversible

thermally triggered lubrication behavior. In contrast, for the continuous temperature-ramp tests in Figs. S3 and S4, the sample remained mounted on the tribometer and was heated in situ using a silicone rubber heating pad during continuous sliding, while the friction coefficient and temperature were recorded simultaneously.

3 Results and discussion

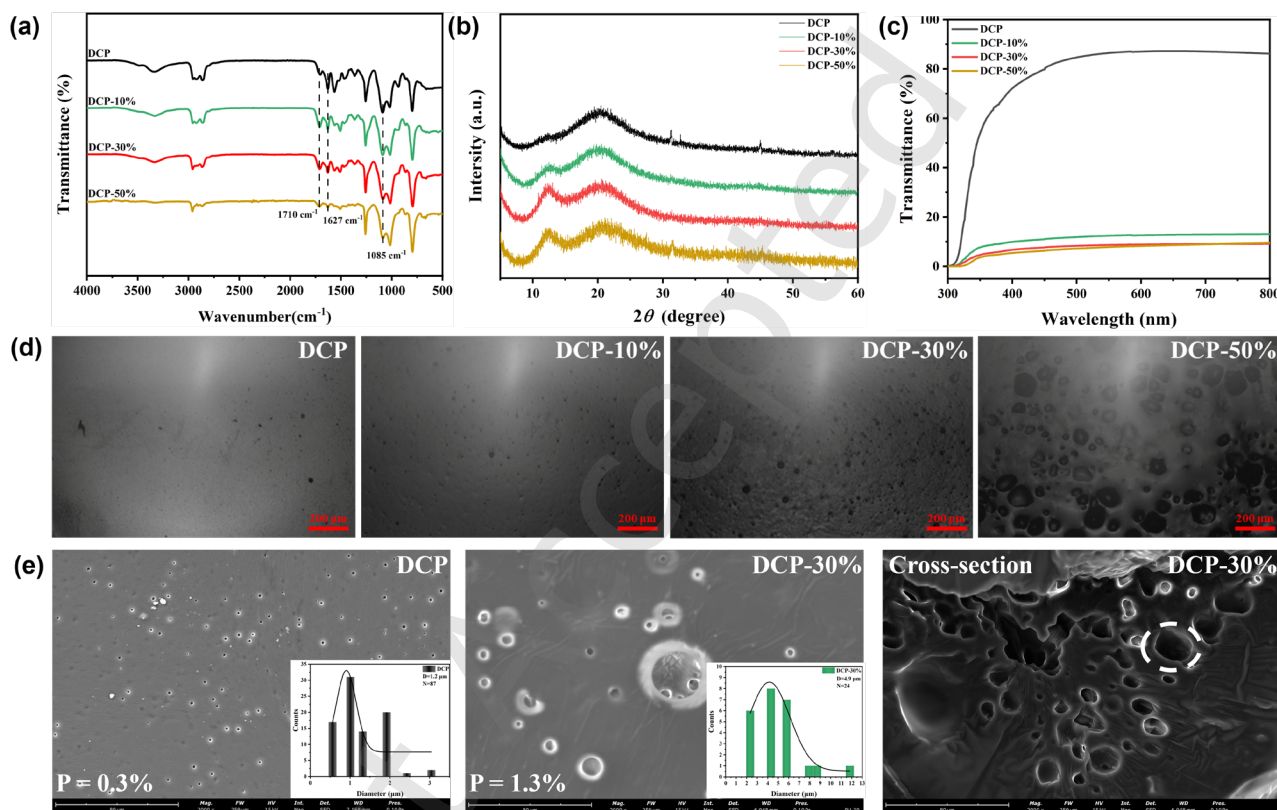


Fig. 2 Fabricated adaptive lubricating oleogel samples by synergistically combining gradient-wettability materials with a dynamic covalent cross-linked network. (a) FTIR spectra. (b) XRD patterns. (c) UV-Vis spectra. (d) Optical micrographs at different volume fractions. (e) Surface and cross-sectional micro-nano pore morphology; Inset: pore size distribution.

To elucidate the origin of friction regulation in the oleogel system, we first examined its structure, thermal response, surface/interfacial properties, and mechanical behavior, as these factors collectively determine the contact state and lubrication behavior during sliding. To verify the chemical structure of the successfully synthesized dynamic covalent cross-linked network oleogel, FTIR was employed to analyze its chemical composition (Fig. 2a). All four samples exhibited

stretching vibration peaks corresponding to urethane and urea groups, detected at 1710 cm^{-1} and 1627 cm^{-1} , respectively, along with a characteristic absorption peak at 1085 cm^{-1} attributed to Si–O–Si bonds in both PDMS segments and the silicone oil backbone. In addition, the absence of signals for isocyanate groups further corroborated the correctness of the oleogel's chemical structure. X-ray diffraction (XRD) analysis was used to examine the crystalline behavior of the dynamic cross-linked polymer DCP and oleogels with varying volume fractions of silicone oil (Fig. 2b). A broad peak observed at 20.7° confirmed the amorphous nature of the DCP sample; upon the introduction of silicone oil, a broad diffraction peak at 12.5° emerged, corresponding to PDMS. The optical transparency of the four samples was evaluated by UV-Vis spectroscopy (Fig. 2c). The results showed that DCP exhibited glass-like transparency, with a transmittance of 86.5% at 550 nm within the visible range, which further verified that the fabricated strategy combining dynamic cross-linkers with gradient-wettability materials induced no significant phase separation, allowing hydrophilic and hydrophobic components to be uniformly distributed throughout the material. However, with the incorporation of silicone oil, the transmittance of the three other samples decreased to 12.4%, 8.7%, and 7.8%, representing a reduction of 85.7%–90.9%. This indicates that silicone oil and the DCP polymer are intrinsically immiscible, and that the locking effect of the dynamic cross-linked network, together with the affinity between PDMS segments in the polymer matrix and silicone oil, firmly anchors the silicone oil within the material.

Optical microscopy was used to examine the surface morphology of the dynamic covalent polymer DCP and the oleogels DCP-10%, DCP-30%, and DCP-50% (Fig. 2d). The DCP surface is flat and smooth with no discernible heterophase structures, which supports a uniform distribution of hydrophilic and hydrophobic constituents. As the silicone-oil content increases, the surface shows progressively more exudation of the silicone-oil phase. At 10 vol.% and 30 vol.% only a few heterophase domains are visible, indicating that most of the lubricant is encapsulated within the internal cross-linked network. At 50 vol.% pronounced exudation appears, implying that the capacity

of the bulk network to host silicone oil has been reached. Scanning electron microscopy was then used to resolve the microstructure of sample surfaces (Fig. 2e). The DCP surface contains uniformly distributed micro-nano pores. Based on 87 pores measured with Image-Pro Plus 6.0, the minimum diameter is approximately 695 nm, the maximum is approximately 2.25 μm , the mean diameter D is approximately 1.2 μm , and the porosity is approximately 0.3%. The pore-size distribution is approximately normal. In the DCP-30% oleogel, uniformly distributed micrometer-scale pores and through-pore structures are observed. From 24 pores analyzed, the mean diameter D is approximately 4.9 μm , which is an increase of 308.3% compared with the pristine sample, and the porosity P increases to 1.3%. Cross-sectional images of the oleogel further reveal abundant micro-nano through-pores in the interior. These results indicate that introducing the lubricant transforms surface micro-nano pores into micrometer-scale pores, increases both pore size and porosity, and that the internal through-pore network provides substantial storage volume for the lubricant.

Under a design in which the primary matrix is constructed from a combination of gradient-wettability materials, we propose an innovative surface hydrophilic-hydrophobic heterogeneous wettability mechanism that enables metastable locking, exudation, and recovery of the lubricating oil through the combined action of thermally induced dissociation in a dynamic cross-linked topological network and the surface micro-pore architecture. As shown in Fig. 3a, the oleogel prepared with gradient-wettability components contains locally low-polarity segments that render a critical volume fraction of liquid silicone oil thermodynamically metastable with the matrix, so no obvious macroscopic solid-liquid phase separation is observed. Upon thermal stimulation at 100 $^{\circ}\text{C}$, when dissociation of the dynamic oxime-urethane linkages exceeds association, disruption of the metastable state decreases the network cross-link density and promotes dynamic migration of the low-surface-energy lubricant, which migrates through the abundant micro-nano pores and enriches at the surface to form a thin lubricating layer. As the temperature decreases, the presence of surface

pores allows the lubricant to be drawn back into the bulk under the driving force of the Laplace pressure P_s .

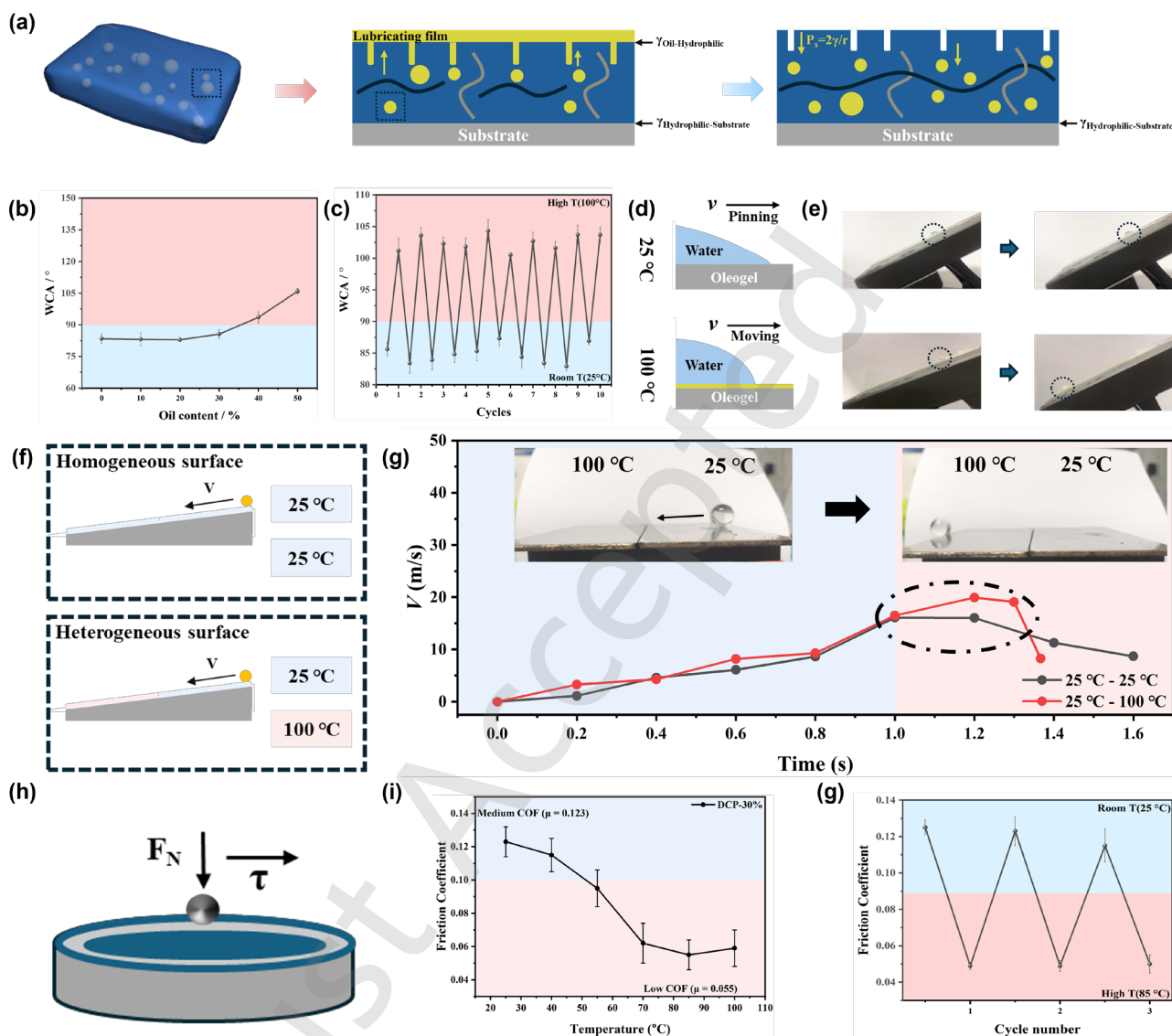


Fig. 3 Schematic illustration of a hydrophilic-to-hydrophobic wettability conversion mechanism via thermally induced dissociation in a dynamically cross-linked topological network. (a) Schematic of the wettability conversion mechanism from hydrophilic to hydrophobic interfacial states. (b) Surface wettability of oleogels at different volume fractions. (c) Schematic of temperature-induced wettability conversion. (d) Schematic of droplet motion at different temperatures. (e) Actual droplet motion on an inclined surface. (f) Schematic of bead motion on surfaces with homogeneous or heterogeneous temperature differences. (g) Velocity plot of bead motion on surfaces with homogeneous or heterogeneous temperature differences. (h) Schematic of the ball-on-disk rotational sliding configuration. (i) Temperature-induced tribological transition of DCP-30%. (g) Cyclic

response of the coefficient of friction under alternating temperatures. (intermittent temperature-switching test).

To further elucidate the correlation between the molecular design of oleogels fabricated via a dynamic covalent topological network and their static surface wettability, we measured WCA of coatings with different silicone-oil contents from 0 vol.% to 50 vol.% (Fig. 3b). The pristine sample (0 vol.%) exhibits a WCA of 83.4° , indicating a hydrophilic state. As the silicone-oil content increases to 30 vol.%, the polymer surface remains hydrophilic. With further increases in volume fraction, the surface becomes hydrophobic, reaching a final WCA of 105.9° . This behavior indicates that at 40 to 50 vol.% the internal dynamic topological network cannot accommodate the excess silicone-oil molecules, which progressively migrate and enrich at the surface due to their low surface energy, thereby producing a hydrophobic state. These results show that the WCA increases progressively with lubricant volume fraction, and that 30 vol.% provides the most suitable internal topological network for locking lubricant molecules during testing. To probe the relationship between compatibility and wettability conversion in a system constructed from a dynamic covalent topological network and gradient-wettability components (Fig. 3c), we selected the 30 vol.% oleogel for subsequent evaluation and conducted ten thermal cycles. After drop-coating the oleogel onto bare glass to form a coating, heating at 100°C for 10 min causes the dissociation rate of dynamic covalent bonds to exceed the association rate. The weakening of cross-link anchoring, combined with the intrinsic low surface energy of silicone oil, drives lubricant migration through micro-nano pores to the surface, where it forms an exuded layer and converts the wettability from hydrophilic (85°) to hydrophobic (102°). After standing at room temperature for 5 to 15 min, Laplace-pressure-driven adsorption associated with the pore architecture returns the surface from hydrophobic to hydrophilic. These observations verify the proposed metastable locking–exudation–recovery mechanism for hydrophilic–hydrophobic heterogeneous wettability.

To clearly demonstrate the wettability conversion characteristics of the oleogel, as shown in Fig. 3d-e, we designed a surface inclined at 30° and placed a 50 μL droplet at the top. At room temperature, the hydrophilic surface of the oleogel pins the droplet at its initial position. After heating at 100 °C for 10 min, the droplet is clearly observed to slide slowly to the bottom. To further highlight the advanced dynamic surface wettability conversion of this material, as shown in Fig. 3f-g, the oleogel was drop-coated onto two 45# steel plates of width 7.5 mm, which were assembled either as a homogeneous pair (both plates unheated) or as a heterogeneous pair (one plate heated, the other unheated). The assembly was placed on a 3° incline. A glass bead of mass 0.61 g served as the probe to monitor motion on the homogeneous and heterogeneous surfaces. On the homogeneous surface, motion on the first unheated plate is similar, with the velocity increasing from 0 m s^{-1} to 16.5 m s^{-1} in about 1 s. On the homogeneous pair the velocity then decreases gradually to 8.7 m s^{-1} , and the bead leaves the plate. On the heterogeneous pair, after crossing the interface between plates across the temperature gradient at 1.0–1.2 s, the velocity surges to 20.0 m s^{-1} , then drops rapidly to 8.3 m s^{-1} by 1.36 s before exiting the field of view. These results indicate that temperature promotes dynamic dissociation of the cross-linked network and exudation of the lubricant phase from surface micro-nano pores, producing a low-surface-energy lubricating layer that causes a marked velocity increase and a faster departure of the bead from the heated region.

To validate switching between hydrophilic and hydrophobic interfacial states under thermal stimulation of the dynamic cross-linked network, we used tribological testing to probe and establish a systems-level relationship linking temperature-triggered association-dissociation in the network, lubricant exudation-recovery, wettability conversion, and switching between medium and low coefficients of friction. As shown in Fig. 3h, a ball-on-disk rotational configuration was used with a normal load of 1 N (20.9 MPa) and a speed of 100 r min^{-1} . Temperature-responsive tribology was further evaluated using the DCP-30% sample (Fig. 3i and Fig. S1). As temperature increases, the average coefficient decreases from 0.123 at room temperature and reaches a minimum at 85 °C with

$\mu = 0.055$. When the treatment temperature is raised to 100 °C, the coefficient increases to 0.064. Thus, the system exhibits a medium-friction viscous state of approximately 0.123 at room temperature. Upon heating, the temperature responsiveness of the dynamic network causes dissociation to exceed association, the encapsulated lubricant transitions from a locked to a random-mobility state, and exudes through surface micro-nano pores to form an oil film, converting solid–solid friction to solid–liquid friction and yielding a low-friction hydrophobic-slip state with $\mu = 0.055$. On the other hand, the relationship between surface wettability and the friction coefficient under temperature variation was verified (Fig. S2). On the other hand, the stability of multiple friction switching events under intermittent friction conditions was verified. As shown in Fig. 3g, during the three test cycles, the coefficient of friction at 25 °C remained stable within the range of 0.115–0.125. With increasing cycle number, the friction coefficient decreased slightly, indicating that the reabsorption of part of the lubricant medium may have been somewhat suppressed. In contrast, at 85 °C, the coefficient of friction remained stable within the range of 0.049–0.051, demonstrating that the lubricant medium still maintained a stable friction regime during cyclic testing after the interval period.

To further validate the tunable range and response time of friction regulation within different temperature windows, a series of customized stepwise heating profiles were designed to evaluate the thermal response behavior of the oleogel system. The dynamic frictional response to temperature input and its programmability were systematically assessed. To ensure comparability among different temperature intervals, all temperature-control experiments were conducted with a 5 or 10 min running-in stage at room temperature (25 °C) to establish a stable initial friction state, and the heating program was initiated at $t = 5$ or 10 min. First, under a customized stepwise heating sequence (25 → 40 → 55 → 70 → 85 → 100 °C), the friction coefficient $\mu(t)$ exhibited a reproducible “transition–stabilization” behavior in response to each temperature step, demonstrating that temperature can serve as an external stimulus to modulate the friction coefficient in the time domain

(Fig. S3). Second, to quantitatively compare the regulation efficiency at different target temperatures, single-step heating experiments ($25\text{ }^{\circ}\text{C} \rightarrow T$) were performed to extract the steady-state friction coefficient and response time for each temperature interval (Fig. S4). The response time Δt was defined as the duration from the onset of heating to the point at which the friction coefficient reached and remained within the vicinity of the target steady-state level (indicated by the red markers in Fig. S4). The results reveal a pronounced and quantifiable frictional regulation upon heating. When the temperature was increased from $25\text{ }^{\circ}\text{C}$ to 40 , 55 , 70 , 85 , and $100\text{ }^{\circ}\text{C}$, the steady-state friction coefficients stabilized at 0.105 , 0.088 , 0.066 , 0.055 , and 0.069 (evolution of the coefficient of friction $\Delta\mu = 0.027$, 0.037 , 0.06 , 0.07 , and 0.05), corresponding to reductions of approximately 20.5% , 29.6% , 47.6% , 56.0% , and 42.0% relative to the initial room-temperature steady state. The associated Δt were 3.6 , 4.2 , 5.9 , 9.7 , and 12.5 min, respectively. Overall, higher target temperatures require longer times for the system to reach a new steady state, reflecting the kinetics of thermal equilibration and interfacial state reconstruction. Notably, the lowest steady-state friction ($\mu \approx 0.055$) is achieved at $85\text{ }^{\circ}\text{C}$, whereas a slight increase in friction is observed at $100\text{ }^{\circ}\text{C}$ ($\mu \approx 0.069$), indicating that the effect of temperature on friction is non-monotonic. This behavior may arise from a competition between enhanced lubrication/interfacial film formation and temperature-induced material softening or changes in film stability. Collectively, the dynamic and steady-state results confirm that the present system exhibits a well-defined, reproducible, and temperature-programmable friction coefficient under thermal stimulation.

In addition, optical microscopy corroborates thermally induced exudation and capillary reabsorption of the lubricant phase (Fig. S5 and Fig. S6). A square-marked region on the oleogel surface was fixed for observation. DCP shows no obvious change after heat treatment. The initial oleogel surface is smooth with no discernible heterophase structures. After heating at $100\text{ }^{\circ}\text{C}$ for 10 min, micro-nano lubricant droplets appear, indicating controllable exudation of the low-surface-energy lubricant driven by thermally activated dissociation of dynamic covalent bonds. After

standing at room temperature for a period of time, no circular lubricant heterophase is observed on the surface, demonstrating recovery of the lubricant mediated by Ps associated with micro-nano pores and by affinity with low-surface-energy PDMS segments in the bulk. Furthermore, exudation during five heat-cool cycles was monitored (Fig. S7). Taken together, the two dynamic wettability tests and optical microscopy jointly validate the proposed surface hydrophilic-hydrophobic heterogeneous wettability mechanism based on metastable locking, exudation, and recovery of the lubricant.

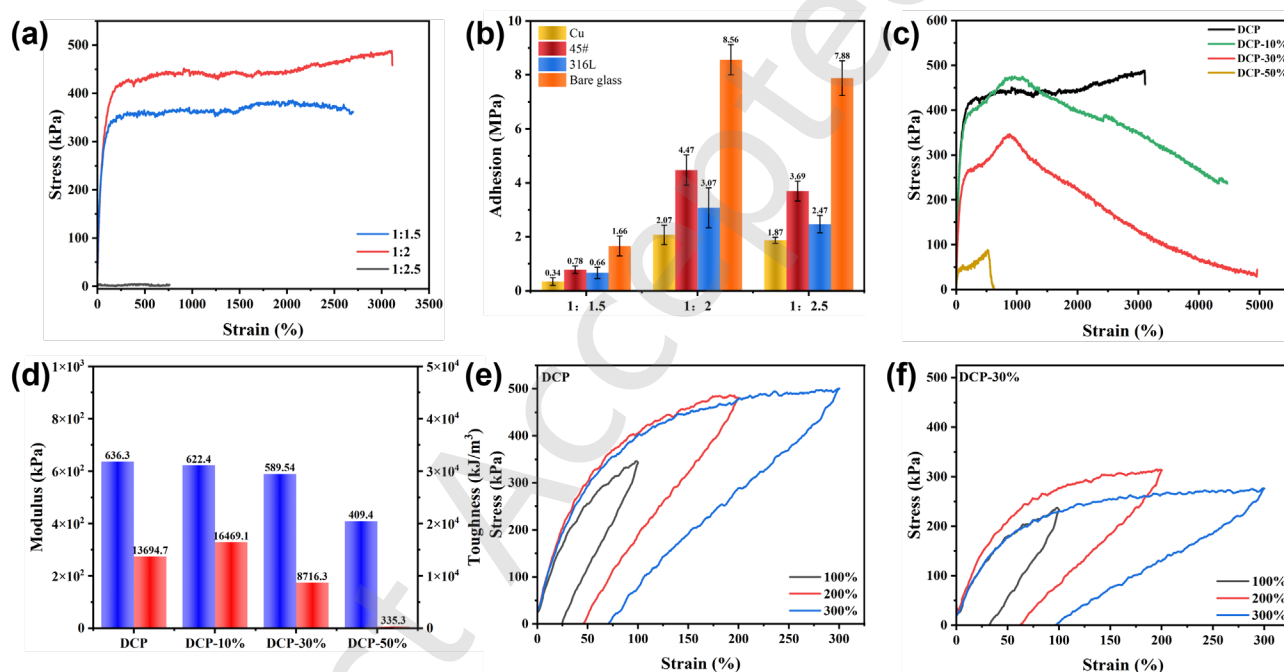


Fig. 4 Mechanical properties of the dynamic cross-linked polymer and the corresponding oleogels. (a) Stress-strain curves of DCPs with different cross-link densities. (b) Interfacial adhesion strength of DCPs with different cross-link densities. (c) Stress-strain curves of oleogels at different volume fractions. (d) Modulus and toughness of oleogels at different volume fractions. (e) Cyclic tensile curves of DCP. (f) Cyclic tensile curves of DCP-30%.

To investigate how the molar content of the DAG influences the mechanical properties of DCP, we selected appropriate cross-link densities to prepare oleogels and characterize their performance. As shown in Fig. 4a and Fig. S8, the molar ratio of total soft-segment hydroxyl groups to DAG was set to 1:1.5, 1:2, and 1:2.5. At 1:1.5, the ultimate stress is approximately 4.9 ± 0.15 kPa and the ultimate

strain is approximately $761.8 \pm 31\%$, with a corresponding modulus and toughness of only 0.96 kPa and 19.5 kJ m^{-3} , indicating that the viscous component dominates the viscoelastic response. At 1:2, the ultimate stress increases to $488.2 \pm 46 \text{ kPa}$ and the ultimate strain to $3106.9 \pm 92\%$, while the modulus and toughness rise to 636.3 kPa and $13694.7 \text{ kJ m}^{-3}$. When the ratio is further raised to 1:2.5, the ultimate stress and strain decrease to $384.1 \pm 51 \text{ kPa}$ and $2697.2 \pm 74\%$, and the modulus and toughness decline to 596.5 kPa and 9736.2 kJ m^{-3} . Consistency of chemical structure and crystalline behavior across the three samples was also verified (Fig. S9). These results indicate that both insufficient and excessive cross-linker contents are detrimental to the mechanical performance of the dynamic polymer, and that a 1:2 molar ratio is optimal.

To further elucidate how varying the molar introduction of the dynamic cross-linker affects the interfacial adhesion of the pristine DCP, oleogels were prepared at appropriate cross-link densities and their performance was characterized (Fig. 4b). The molar ratios of total soft-segment hydroxyl groups to DAG were set to 1:1, 1:1.5, 1:2, and 1:2.5. At 1:1 the cross-link density is too low, the polymer remains molten, and solidification cannot be achieved. With increasing cross-linker content, the interfacial adhesion strength of DCP on bare glass first increases and then decreases, measuring 1.66 MPa, 8.56 MPa, and 7.88 MPa. Adhesion on metallic substrates (copper, 45# steel, and 316L stainless steel) was also assessed. On 45# steel, which shows comparatively better adhesion among the metals, the strengths are 0.78 MPa, 4.47 MPa, and 3.69 MPa for the three cross-linked conditions, consistent with the trend observed on glass. These results align with the tensile data: low cross-link density yields weak mechanical performance, increasing the density to 1:2 produces optimal interfacial adhesion, and further increases reduce adhesion. Accordingly, subsequent studies employed samples with a moderate cross-linker loading at a 1:2 ratio.

Building on the above results, a 1:2 molar ratio was selected for subsequent experiments. DCP-vol.% were prepared by adding 10 vol.%, 30 vol.% and 50 vol.% silicone oil, and tensile tests were performed (Fig. 4c-d). Compared with DCP, the silicone-oil-containing samples exhibit pronounced

yielding. For DCP-10% the ultimate stress and strain are 474.3 ± 57 kPa and $4468.1 \pm 127\%$, with a modulus and toughness of 622.4 kPa and $16469.1 \text{ kJ m}^{-3}$, respectively; the modulus decreases slightly while the toughness increases by 20.3%. For DCP-30% the ultimate stress and strain are 345.7 ± 37 kPa and $4956.7 \pm 197\%$, with a modulus and toughness of 589.5 kPa and 8716.3 kJ m^{-3} ; the modulus decreases by 7.4% and the toughness decreases by 36.4%. For DCP-50% the ultimate stress and strain are 86.9 ± 16.2 kPa and $619.8 \pm 82\%$, and the modulus and toughness decrease by 35.7% and 97.6%, respectively. These results indicate a clear plasticization effect upon silicone-oil incorporation, while the gradient-wettability materials design attenuates what would otherwise be a severe loss of mechanical performance. The 30 vol.% oleogel was selected for subsequent comparative validation.

Cyclic tensile tests were conducted on DCP and on the optimized DCP-30% at fixed maximum strains of 100%, 200%, and 300% (Fig. 4e-f). For DCP the residual strains at 100%, 200%, and 300% are 24.7%, 46.2%, and 70.8%, respectively. For DCP-30% the residual strains at 100%, 200%, and 300% are 31.9%, 63.6%, and 96.8%, respectively. DCP-30% exhibits larger residual strain and a smaller hysteresis loop than DCP at all three strain levels. This is mainly attributed to plasticization by the highly compatible silicone oil, which promotes chain-segment slippage, reduces interchain friction, and effectively lowers energy dissipation.

To further elucidate the lubricant-exudation mechanism underlying the controllable surface wettability transition of DCP and the optimized DCP-30%, we investigated the governing thermodynamics, viscoelastic properties, and stress-relaxation behavior. As shown in Fig. 5a, the glass-transition temperature of DCP prepared at a 1:2 ratio is approximately 23.11 °C, and no distinct endothermic melting peak is observed within the -70 to 100 °C window, indicating an amorphous structure. Upon addition of 30 vol.% silicone oil, the oleogel exhibits a markedly reduced T_g of about -1.49 °C, suggesting that silicone oil within the dynamic covalent topological network weakens

intersegmental interactions; plasticization together with enhanced chain flexibility synergistically depresses T_g . In addition, the thermodynamic stability of the corresponding materials was verified (Fig. S10).

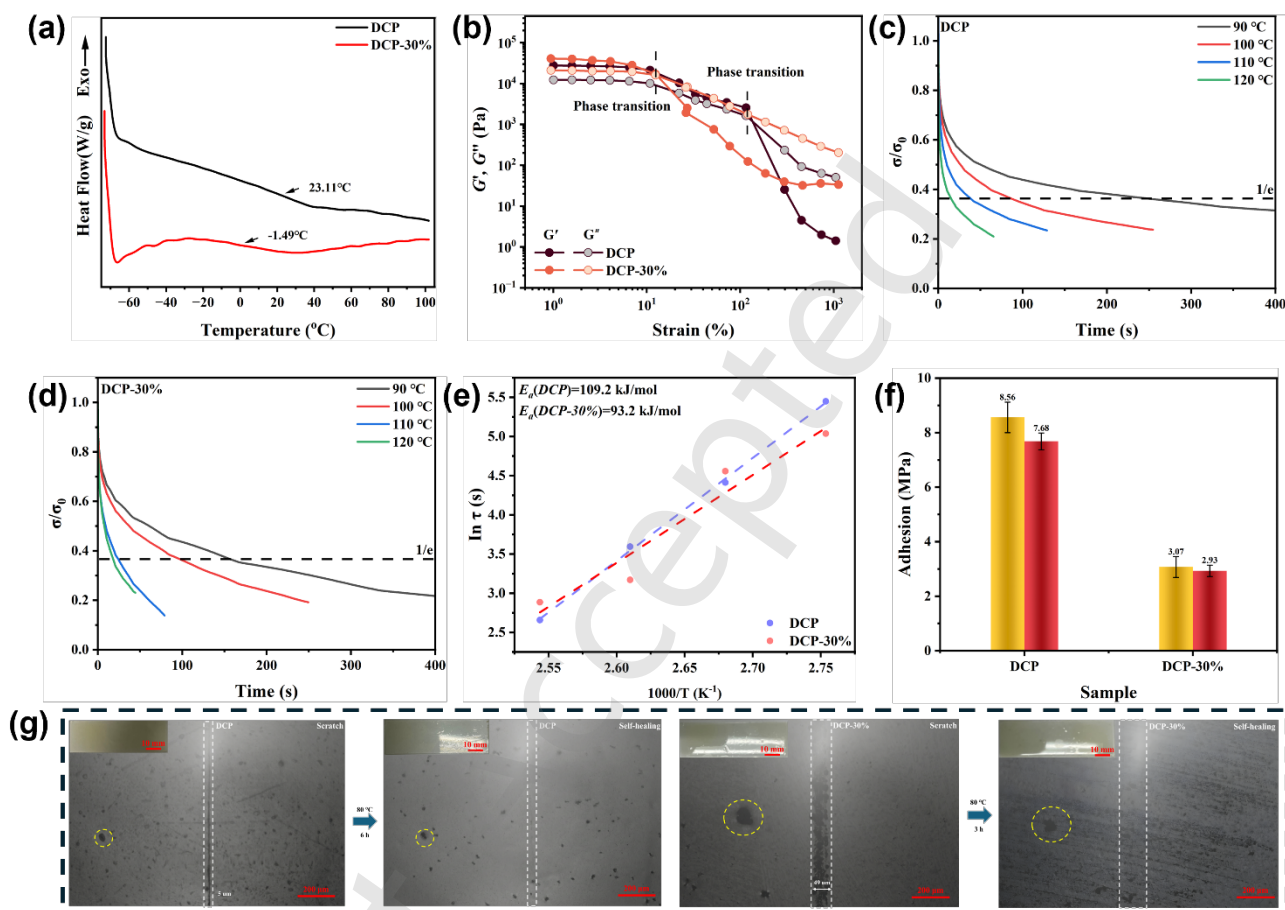


Fig. 5 Thermodynamic and rheological relaxation characteristics of the samples. (a) DSC curves of DCP and DCP-30%. (b) Rheological behavior of DCP and DCP-30%. (c) Stress-relaxation curves of DCP at different temperatures. (d) Stress-relaxation curves of DCP-30% at different temperatures. (e) Activation energy obtained by fitting the Arrhenius equation. (f) Interfacial adhesion strength before and after self-healing. (g) Optical micrographs and digital photographs documenting the self-healing process.

Owing to the association-dissociation dynamics of the covalent bonds, strain-sweep rheology was used to resolve the viscoelastic response. As shown in Fig. 5b, the storage and loss moduli of DCP cross at increasing shear strain ($G' = G''$), marking a gel-to-sol transition in which elastic behavior gives way to viscous behavior. In contrast, DCP-30% exhibits the $G' = G''$ crossover at

much lower strain, indicating that the presence of the lubricant phase facilitates dynamic dissociation of the cross-linked network. This arises because the lubricant increases the viscous contribution and lowers the effective cross-link density, thereby weakening the elastic network. Rheology further indicates controllable exudation of the lubricant via dynamic association–dissociation through the network, consistent with wettability conversion and an intelligent self-lubricating mode. Complementary stress-relaxation tests corroborate the dynamics imparted by oxime-urethane linkages (Fig. 5c-e). For both DCP and DCP-30%, the relaxation modulus decays over time and the characteristic relaxation time shortens as temperature increases from 90 °C to 120 °C; DCP-30% relaxes faster due to the lubricant phase. Activation energies for both systems were obtained by fitting the Arrhenius equation.

$$\tau = \tau_0 \exp\left(\frac{E_a}{RT}\right)$$

Here τ denotes the relaxation time, τ_0 is the pre-exponential factor, E_a is the activation energy in kJ mol⁻¹, R is the gas constant with a value of 8.314 J mol⁻¹ K⁻¹, and T is the absolute temperature in kelvin.

The relaxation activation energies of DCP and DCP-30% are 109.2 kJ mol⁻¹ and 93.2 kJ mol⁻¹, respectively, indicating that introduction of the lubricant phase lowers the activation barrier for relaxation and facilitates dynamic dissociation of the dynamic covalent cross-linkers.

To macroscopically demonstrate the dynamic dissociation-association of oxime-urethane linkages, we investigated the self-healing behavior of the dynamic covalent polymer DCP and the corresponding oleogel DCP-30%. As shown in Fig. 5g, optical microscopy reveals a 5 μm wide scratch on DCP, and the inset shows the macroscopic scar. After heat treatment at 80 °C for 6 h, the surface scratch disappears, achieving self-healing. For DCP-30%, a scratch approximately 49 μm in width fully heals after 3 h at 80 °C. These observations indicate that introduction of the liquid silicone-oil phase facilitates temperature-triggered association-dissociation of oxime-urethane bonds, enabling macroscopic surface self-healing, in agreement with the rheological relaxation behavior

described above. We further quantified healing efficiency via interfacial adhesion before and after healing (Fig. 5f). For DCP on bare glass, the initial interfacial adhesion strength is about 8.56 MPa and recovers to 7.68 MPa after healing, corresponding to a healing efficiency of approximately 90%. For DCP-30%, the initial strength is reduced to 3.07 MPa due to the presence of liquid silicone oil and recovers to 2.93 MPa after healing, giving an efficiency of approximately 95%. These results show that DCP-30% heals faster and more effectively than DCP. Consistency of chemical structure before and after healing was verified (Fig. S11), indicating that the thermal healing protocol does not compromise the intrinsic physicochemical properties.

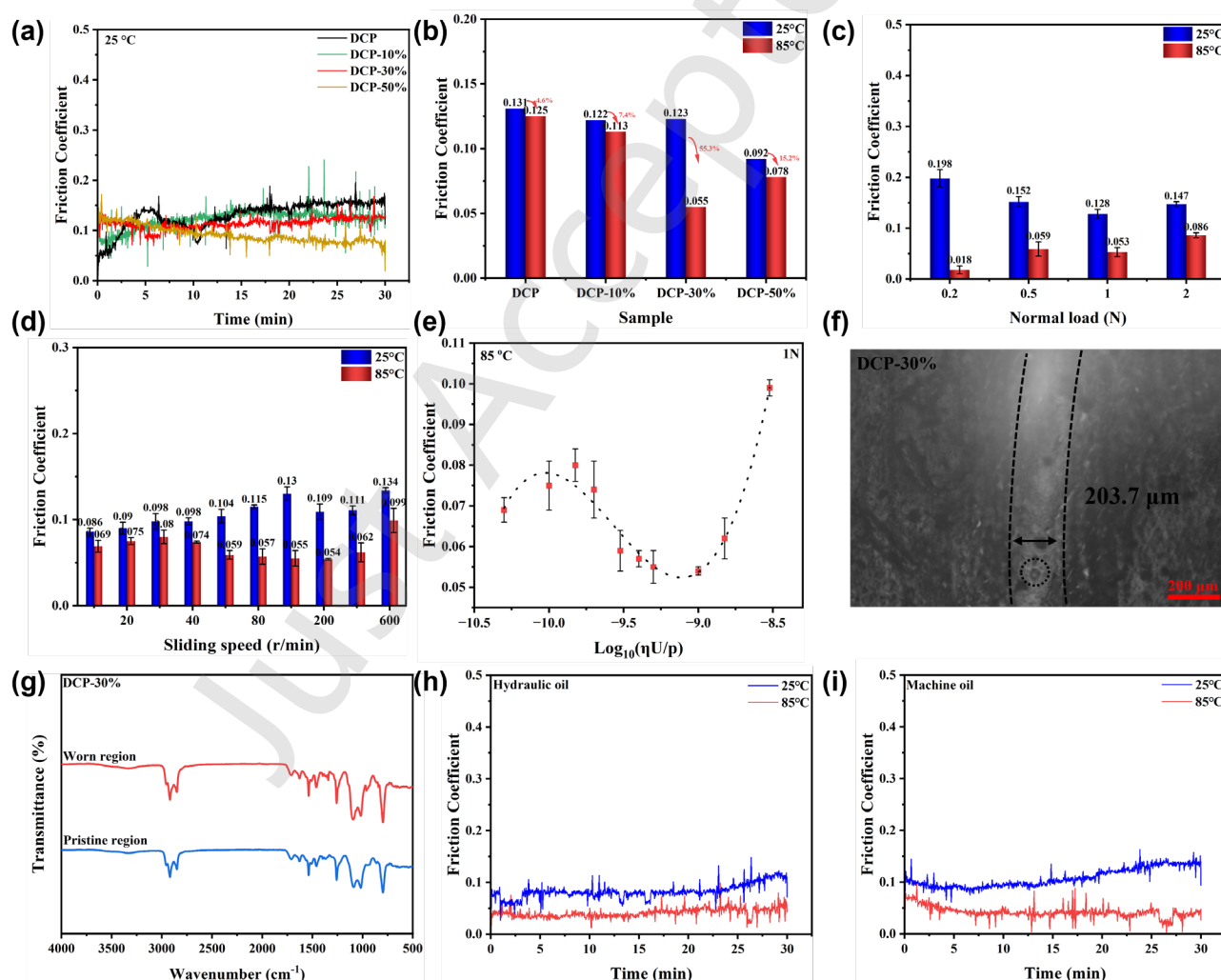


Fig. 6 Tribological behavior of dynamically cross-linked oleogels exhibiting thermally stimulated switching of the intelligent self-lubricating state. (a) Coefficients of friction at 25 °C for different volume fractions. (b) Temperature-induced differences in the coefficient of friction among samples

with different volume fractions. (c) Friction coefficient of DCP-30% under different loads. (d) Friction coefficient of DCP-30% at different sliding speeds. (e) Stribeck curves of the oleogel surface after heat treatment at 85 °C. (f) Wear-track images of DCP-30%. (g) FTIR spectra from the wear zone and the unworn zone of DCP-30%. (h) Temperature-induced tribological behavior in a hydraulic oil medium. (i) Temperature-induced tribological behavior in a machine oil medium.

Based on the above structural and interfacial characterizations, we further evaluated the friction-regulation behavior of the oleogel under different external conditions. Fig. 6a summarizes room-temperature tribology at 25 °C for DCP and oleogels with different lubricant volume fractions. DCP without silicone-oil lubricant exhibits an average coefficient of friction of approximately 0.131 with substantial fluctuations, attributed to poor lubrication arising from uniformly distributed micro-nano pores. With increasing lubricant content, DCP-10% and DCP-30% show average coefficients of 0.122 and 0.123 at 25 °C with improved stability, representing only 6.9% and 6.1% reductions relative to DCP. This indicates that 10% to 30% lubricant is fully encapsulated within the dynamic network, consistent with the wettability results. When the lubricant fraction reaches 50%, DCP-50% shows an average coefficient of approximately 0.092 at 25 °C, a 29.8% reduction, confirming that the network can no longer accommodate 50% silicone oil and that a surface oil film reduces friction.

As shown in Fig. 6b and Fig. S12-13, comparison across volume fractions revealed that DCP without lubricant showed only a 4.6% decrease in friction, indicating no dual-state switching (Fig. S12). In other words, the polymer is only weakly affected within the customized temperature range of 25-100 °C. DCP-10% decreased by only 7.4%, indicating insufficient lubricant to exude from the bulk. DCP-30% showed excellent temperature responsiveness, switching from the medium-friction state to a low-friction state with a 55.3% reduction, thereby achieving dual lubrication states governed by wettability conversion. For DCP-50%, the presence of a pre-existing surface oil film resulted in only a 15.2% decrease in friction upon heating. These results demonstrate that the cooperative ratio between lubricant content and the dynamic cross-linked network is the key factor governing the thermally induced transition to a metastable intelligent self-lubricating state.

To further strengthen the interpretation of the friction mechanism, the friction coefficient variation of the DCP-30% sample was systematically compared under different normal loads and sliding speeds. Fig. 6c shows the load-scanning results (0.2-2 N) at a fixed speed of 100 rpm, while Fig. 6d presents the speed-scanning results (10-600 rpm) under a fixed load of 1 N. Overall, the friction coefficient at 85 °C is significantly lower than that at 25 °C, accompanied by reduced fluctuations, suggesting that interfacial shear mechanisms are substantially altered upon heating.

Under the load-scanning mode at 100 rpm (Fig. 6c), the friction coefficient at 25 °C decreases as the normal load increases from 0.2 N to 1 N, followed by a slight increase at 2 N. This non-monotonic dependence reflects a higher load sensitivity of the contact state at room temperature, where increasing load modifies the real contact area and interfacial shear stress, leading to a complex friction response. In comparison, the friction coefficient at 85 °C remains at a low level over 0.2-2 N and shows only a weak dependence on load, implying a relatively stable silicone-oil-assisted lubrication contribution even when the lubricant layer is compressed under higher contact pressure. Collectively, these results demonstrate that increasing temperature can significantly reduce friction and enhance stability: at 25 °C the friction is mainly governed by solid contact/adhesive shear contributions, whereas at 85 °C the participation of silicone oil is enhanced and becomes the dominant interfacial shear pathway. On the other hand, under a low load (0.2 N), the average μ of the high-temperature-induced surface decreased from 0.198 at room temperature to 0.018 (a reduction of 91%). Within the intermediate load range (0.5-1 N), the average μ decreased from 0.14 to 0.056 (a reduction of 60%). Under a high load (2 N), the average μ decreased from 0.147 to 0.086 (a reduction of 41%). These results indicate that the temperature-induced variation in the coefficient of friction decreases monotonically with increasing load, suggesting a transition in the contact mode between the friction pair and the oleogel.

Under the speed-scanning mode at 1 N (Fig. 6d), the friction coefficient at 25 °C remains relatively high and exhibits more pronounced fluctuations in the low-to-intermediate speed range.

With increasing rotational speed, the friction coefficient still shows an increasing tendency or a larger scatter. This behavior indicates that at room temperature the migration and replenishment of silicone oil at the contact interface are limited, and the contact is more likely dominated by solid/viscoelastic surface interactions and local adhesive shear, resulting in a stronger speed dependence and poorer friction stability. By contrast, at 85 °C the friction coefficient is markedly reduced and becomes smoother, displaying a decreasing or plateau-like trend with increasing speed. This suggests that elevated temperature facilitates oil enrichment within the contact region and promotes the formation of a lubrication-mediated low-shear interface, where the dominant shear occurs within the silicone-oil-rich layer, thereby lowering friction and suppressing fluctuations. On the other hand, when the sliding speed was ≤ 40 rpm, the average μ of the high-temperature-induced surface decreased from 0.093 at room temperature to 0.075 (a reduction of approximately 19%), which is mainly attributed to the viscous effect of the polymer surface at low speeds. When the sliding speed was in the intermediate range (60-400 rpm), the average μ decreased from 0.114 to 0.057 (a reduction of 50%), primarily because a continuous lubricating oil film could be readily formed on the polymer surface at moderate speeds. When the sliding speed increased to 600 rpm, the average μ decreased from 0.134 to 0.099 (a reduction of 26%), which is mainly attributed to the oil film being expelled at high speeds, making it difficult to maintain a continuous lubricating film.

To further and systematically investigate the effect of temperature on the evolution of interfacial lubrication states, Stribeck curves [59] were constructed at a fixed load of 1 N for both 25 °C and 85 °C by plotting the friction coefficient μ as a function of the dimensionless parameter $\log_{10}(\eta U / p)$. Here, the sliding speed was calculated as $U = 2\pi r \cdot rpm / 60$; the nominal pressure was estimated by $p = F / A$, where the contact area A was obtained from the wear scar geometry; and the silicone oil viscosity at different temperatures was measured using a viscometer (Fig. S14). Therefore, the difference in the horizontal axis between the two curves is mainly determined by the combined effects of viscosity and speed. As shown in Fig. 6e, Fig. S15 and Table S1, the Stribeck

curves at the two temperatures exhibit distinct characteristics.

At 85 °C, the overall friction level is substantially lower: μ first decreases with increasing $\log_{10}(\eta U / p)$ and reaches a minimum at around 200 rpm ($\mu \approx 0.054$), followed by a rise at higher speeds ($\mu \approx 0.099$ at 600 rpm). This trend implies a progressive transition from boundary/mixed lubrication toward a regime where the lubricant increasingly contributes to load sharing and interfacial shear, with the minimum corresponding to the most effective lubrication condition. The upturn at the high-speed end is more likely attributed to enhanced viscous shear and drag contributions of the lubricant. In contrast, at 25 °C the friction coefficient is higher and more speed-sensitive: μ increases from approximately 0.086 to 0.13 over 10-100 rpm, shows a moderate drop at 200 rpm ($\mu \approx 0.109$), and then increases again toward higher speeds ($\mu \approx 0.134$ at 600 rpm). Within the investigated speed window, the 25 °C curve does not exhibit a distinct low-friction plateau or well-defined minimum as observed at 85 °C, which is more consistent with boundary lubrication or oil-starved conditions.

In summary, these comparisons indicate that heating to 85 °C promotes the formation of a stable lubrication-mediated shear layer, leading to significantly reduced friction and a more typical Stribeck-like minimum. At 25 °C, the interface is more likely governed by boundary/oil-deficient lubrication, where friction remains higher and increases more readily with speed due to combined adhesive shear and viscous dissipation. This observation is consistent with the proposed mechanism that elevated temperature facilitates silicone-oil migration and replenishment at the interface, thereby enhancing lubrication effectiveness.

Using the wear track of the optimized DCP-30% as the basis for analysis (Fig. 6f), a track width of approximately 203.7 μm was observed, and numerous micrometer-scale pores are exposed on the worn surface, indicating ample internal storage space for the lubricant. In parallel, FTIR spectroscopy was used to compare the chemistry of the wear and unworn regions (Fig. 6g). No discernible changes in characteristic absorption peaks were detected, which strongly supports the

chemical stability of the oleogel under tribological conditions. We further examined the generality of state switching using different lubricants (Fig. 6h-i, Table S1). With hydraulic oil as the lubricant, the oleogel shows a room-temperature coefficient of friction of about 0.083 with good stability; upon heating to 85 °C, the average coefficient decreases to about 0.041, a reduction of 50.6%, while stability remains excellent. With machine oil as the lubricant, the room-temperature coefficient is about 0.108 and increases gradually over time; after treatment at 85 °C, it decreases to 0.042, a reduction of 61.1%, and the frictional stability improves. We also investigated how temperature responsiveness affects surface wettability in these two lubricant systems (Fig. S16). These results verify the broad applicability of oleogels prepared by introducing different lubricants into the dynamic cross-linked polymer network. In summary, oleogels that match the lubricant with the dynamic network can, under thermal stimulation, realize switchable hydrophilic and hydrophobic interfacial states as an intelligent self-lubrication mode, which is promising for intelligent control of tunable tribological behavior in robotic joints that transition between solid–solid and solid–liquid contact modes.

4 Conclusions

Inspired by the friction-induced secretion of polymeric mucus from earthworm skin during soil interaction, which enables low friction and self-lubrication, an oleogel that operates in an intelligent self-lubrication mode and undergoes a thermally triggered transition from a hydrophilic weak-adhesion state to a hydrophobic slip state was fabricated by rationally matching a purpose-designed gradient-wettability materials set (polyethylene glycol, polytetrahydrofuran, and hydroxyl-terminated polydimethylsiloxane) with lubricants (silicone oil, hydraulic oil, and machine oil) and by leveraging the thermally induced dissociation and association of a dynamic oxime-urethane cross-linked network.

The dynamic covalent polymer exhibited outstanding mechanical performance, including an ultra-high ultimate strain of $3106.9 \pm 92\%$, a toughness of 13.7 MJ m^{-3} , and robust interfacial adhesion

strength of 8.56 MPa. The optimized oleogel, owing to the incorporated lubricant phase, attained an ultra-high ultimate strain of $4956.7 \pm 197\%$ together with a reduced toughness of 8.7 MJ m^{-3} . Thermodynamic behavior in concert with rheological measurements revealed a sol-gel transition in oleogels bearing dynamic cross-linking sites and demonstrated an excellent thermally driven self-healing efficiency of 95%. The synergy between thermally triggered bond dissociation and association and the abundant micro-nano pore architecture observed at the surface and in the bulk elucidated a wettability-conversion mechanism in which the lubricant is locked, exuded, and recovered, switching the surface from an effectively hydrophilic to a hydrophobic interfacial state. Microscopic observations and wettability assessments (hydrophilic-to-hydrophobic surface transition) captured over ten thermally responsive cycles of lubricant exudation and recovery, and the motion of water droplets and glass beads on homogeneous and heterogeneous surfaces at room and elevated temperatures were investigated, showing velocity increases from 16.5 m s^{-1} to 20.0 m s^{-1} . Based on the above wettability-conversion mechanism, a bio-inspired oleogel exhibiting an intelligent self-lubrication mode was advanced and its temperature-responsive tribological behavior was investigated. At room temperature the oleogel surface showed a medium coefficient of friction ($\mu = 0.123$) under solid-solid contact. After heat treatment at 85°C the surface attained a low coefficient ($\mu = 0.055$) with a contact-mode switch to solid-liquid contact. Studies with different lubricants, including hydraulic oil and machine oil validated the generality of this wettability-driven, contact-mode-switchable self-lubrication. This work is expected to provide new avenues for the design and prospective engineering applications of intelligent robotic joints and motion and transmission components, as well as intelligent self-lubricating materials.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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