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Thermoresponsive Ionogels with Highly Switchable Adhesion

Properties Based on Dynamic Interchain Interactions

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Abstract

Adhesives capable of rapidly and reversibly switching between adhesive and non-adhesive states are desirable for a wide range of applications, including medical adhesives, robotic systems, bioelectronics, and scalable microassembly. In this study, we reported a poly(ionic liquid)-based ionogel that exhibited switchable adhesion to a variety of substrates in response to a mild thermal stimulus. This switchable adhesive behavior arose from the exposure of functional groups on ionogel surfaces caused by the temperature-induced collapse of polymer networks. The adhesion strengths of the ionogel were strongly temperature-dependent and exhibited a large

1 tunable range, with an approximately 60-fold adhesion difference between low-
2 temperature (1.4 kPa @ 0°C) and high-temperature (89 kPa @ 40°C) conditions.
3 Using the ionogel as switchable adhesives on moving parts, three different robotic
4 systems, including two climbing robots and a vertical transport tower, were
5 implemented for application demonstrations. The tethered climbing robots are
6 capable of climbing on various types of vertical walls, including ceramics, metal,
7 wood, glass, plastic, and porous substrates. Locomotion on inverted walls was also
8 demonstrated. In addition, stable vertical climbing with a payload that is 1.5 times
9 greater than the robot's weight was achieved. This study provides a facile and
10 versatile strategy to endow ionogels with tunable, switchable, and dynamic adhesion
11 performance that can expand the applicability of next-generation smart devices
12 across various domains.

13 **Introduction**

14 The development of adhesive technologies is instrumental for the robust and
15 durable assembly of diverse materials in modern society. An adhesive is a substance
16 that promotes interfacial bonding and resists separation¹. Permanent adhesives have
17 long been favored for their mechanical strength and reliability^{2,3}, while switchable
18 adhesives have attracted extensive research interest for their applications in climbing
19 robotics⁴, intelligent gripping⁵, wound dressings⁶, human-machine interfaces⁷,

1 transfer printing⁸, and bioelectronics⁹. A switchable adhesive is a material whose
2 bonding strength can be modulated by external stimuli or varying environmental
3 conditions^{10,11}. Unlike permanent adhesives, switchable adhesives can provide
4 temporary adhesion and controlled debonding on demand, with minimal risk of
5 surface damage.

6 Combining high adhesive strength with pronounced switchability is a
7 fundamental challenge in the design of switchable adhesives. In general, stronger
8 adhesive strengths are correlated with greater detachment difficulty levels, so
9 currently available switchable adhesives are either limited in releasability/reusability,
10 or constrained by low adhesion strength^{12,13}. Moreover, the bonding intensity of an
11 adhesive across different surfaces is governed by both interfacial adhesion and the
12 bulk cohesive strength of the adhesive phase. The prevalent trade-off between
13 cohesion and interfacial adhesion thus constrains the development of high-
14 performance switchable adhesives^{14,15,16}. Previous studies have shown that adhesion
15 switching can be achieved by regulating material cohesion and interfacial
16 interactions.

17 Cohesion-regulated switchable adhesive materials are typically designed to
18 have varying adhesion strengths when their mechanical properties are adjusted^{17,18,19}.
19 However, the majority of these materials encounter significant drawbacks, such as

1 the formation of surface residues after separation, slow switching between adhesive
2 and nonadhesive states, the need for complicated control architectures, and
3 undesirably strong adhesion during detachment. Switchable adhesive materials
4 regulated by interfacial interactions can be classified as exhibiting either covalent
5 adhesion or noncovalent adhesion^{20,21}. Adhesive materials of the covalent type
6 exhibit long-term adhesion stability through covalent crosslinking at the contact
7 interface or via entanglement between the surfaces in contact^{22,23,24}. The formation
8 of strong covalent bonds leads to robust interfacial adhesion but unavoidably requires
9 prolonged separation times and substantial energy inputs for detaching. Noncovalent
10 interfacial adhesion, which is mediated by van der Waals forces, hydrogen bonding,
11 coordination, electrostatic interactions, or cation- π interactions^{25,26,27,28,29,30}, has
12 great potential for the development of switchable adhesive materials with residue-
13 free and rapidly tunable properties, owing to the inherently reversible nature of these
14 interactions with surfaces. Furthermore, noncovalent-type materials are capable of
15 modulating their adhesiveness in response to various stimuli, including pH, pressure,
16 water, and temperature^{31,32,33,34}. These reversible, stimuli-responsive characteristics
17 make noncovalent materials suitable for switchable adhesives in various fields such
18 as sensing, intelligent medicine, robotics, and surface engineering³⁵.

19 Climbing robots have received sustained interest across academia and industries.

1 The key strategies underlying these robots are of two types: the locomotion
2 mechanism and the adhesion mechanism. Animal-inspired locomotion is commonly
3 adopted for climbing, including legged gaits³⁶, linear translation³⁷, inchworm-like
4 motion³⁸, and wheeled mobility³⁹. However, climbing robots that solely rely on
5 locomotion mechanisms can only climb relatively gentle slopes; they face difficulties
6 during inverted climbing unless the substrate has textured surfaces that provide
7 support. Recent studies on climbing robots involving locomotion mechanisms have
8 mainly focused on the integration of additional adhesion mechanisms for improving
9 climbing performance. Adhesion mechanisms are established in climbing robots via
10 several technologies, including vacuum suction^{40,41}, magnetic attachment^{42,43},
11 electrostatic attraction^{44,45}, mechanical gripping^{46,47}, and dry adhesives^{48,49}.
12 Notwithstanding these developments, the production of climbing robots with reliable,
13 switchable adhesion across diverse wall types remains a persistent challenge. For
14 example, climbing robots consisting of vacuum suction mechanisms usually require
15 bulky pneumatic actuation units, and their functionality is limited to climbing rigid,
16 nonporous wet surfaces. Climbing mechanisms that involve electrically controlled
17 adhesives generally depend on the presence of conductive substrates to achieve
18 switchable adhesion or require high-voltage input from an external power supply.
19 Methods involving magnetic adhesion generate strong holding forces, but require

1 ferromagnetic substrates.

2 In this study, we developed a strategy that combines molecular-structure design
3 with network dynamics control to reconcile the cohesion–adhesion trade-off and
4 resolve the incompatibility between strong adhesion and facile detachment, thereby
5 transforming commercial polymers into robust, smart switchable adhesives that
6 achieve both high adhesion and a large switching ratio. Ionogel properties were finely
7 tuned by varying the ionic liquid (IL) composition—specifically, the identities of the
8 constituent cations and anions—leading to an exceptional degree of control over their
9 performance and enabling the rational design of adhesives tailored to specific
10 applications. The resulting adhesive ionogels could swiftly transition between a rigid
11 nonadhesive state and a soft adhesive state within a narrow, physiologically mild
12 temperature window. This capability can facilitate the development of climbing
13 robots functionalized with ionogel pads that, through coordinated temperature
14 control, can achieve switchable adhesion and locomotion on various substrates,
15 including inverted surfaces. Our proposed climbing scheme involving switchable
16 adhesives effectively addresses the critical challenge of adapting to complex,
17 discontinuous, or porous surfaces, thereby substantially broadening the potential
18 capabilities and application scope of climbing robots.

1 **Results and discussion**

2 **Design principle and adhesion mechanism of the ionogels**

3 Conventional ionogels are characterized by uniform crosslinking mediated
4 through reversible physical interactions. Their rheological properties are governed by
5 the interplay between the association energy which facilitates the reformation of
6 interchain crosslinks, and the thermal energy which disrupts interchain associations. At
7 low temperatures, the formation of interchain crosslinks prevails, and the material
8 behaves like a typical gel. At high temperatures, the disruption of interchain
9 associations predominates, which allows for the reptation of entire polymer chains.
10 Hence, the material appears like a sol, and exhibits liquid-like dissipative behavior,
11 which is beneficial for interfacial adaptability during adhesion and leads to a substantial
12 enhancement of adhesive performance. Consequently, by heating and cooling an
13 ionogel so that it switches between the adhesive sol-like state and the non-adhesive gel-
14 like state, the adhesive properties of the ionogel can be reversibly modulated.

15 Our proposed ionogel material features an elastic polymer network formed from 1-
16 vinyl-3-butyylimidazolium tetrafluoroborate (VBIMBF₄, acting as monomers). The
17 ionic liquid 1-butyl-3-methyylimidazolium tetrafluoroborate (BMIMBF₄, acting as the
18 ionic liquid solvent) is embedded within the network, effectively preventing phase
19 separation and ensuring compositional stability. Further, 2,4,6-trimethylbenzoyl

1 diphenylphosphine oxide (TPO) acts as the photoinitiator for photopolymerization.
2 Figure 1a presents the chemicals used to prepare the ionogel precursor solution as well
3 as the polymer network obtained after polymerization. Figure 1b presents a schematic
4 of the photopolymerization process. We used a homogeneous precursor solution
5 consisting of VBIMBF₄, BMIMBF₄, and TPO in this study. Upon UV exposure, the
6 TPO generated free radicals that initiated VBIMBF₄ polymerization (under nitrogen gas
7 within a glovebox), resulting in the formation of poly(VBIMBF₄), a liquid crystalline
8 poly(ionic liquid) (PIL). Owing to the structural similarity between the cation and anion
9 components of PIL and the ionic liquid (IL) BMIMBF₄, the resulting ionogel system
10 exhibited excellent internal compatibility and effectively prevented solvent leakage.

11 A complete description of the precursor formulation is provided in the Methods
12 section. To systematically modulate the adhesive properties of ionogel samples, the
13 mass fraction of the ionic liquids was precisely varied, as detailed in Supplementary
14 Table 1. The resulting ionogels were each labelled ionogel-xx, with “xx” denoting the
15 overall weight percentage (wt%) of the VBIMBF₄ content (i.e., PIL content). Fourier
16 transform infrared (FTIR) spectroscopy analysis was performed to verify the complete
17 polymerization of the VBIMBF₄ monomer. The FTIR spectrum (Supplementary Fig. 1
18 and Supplementary Table 2) showed that the characteristic absorption band at
19 1655 cm⁻¹, attributed to the C=C stretching of the ionic liquid monomer (VBIMBF₄),

1 completely disappeared after polymerization, indicating that the monomer was
2 polymerized completely (Supplementary Fig. 2).

3 Figure 1c presents the proposed temperature-responsive adhesion mechanism for the
4 polymerized ionogels. Upon cooling to below softening temperature (T_s), the
5 equilibrium of electrostatic complexation shifted toward the associated state, and the
6 polymer chains within the ionogel established a physically crosslinked network, thereby
7 reducing molecular mobility as a consequence of intensified interchain interactions.
8 This transition resulted in ionogel stiffening (Supplementary Fig. S3), which
9 substantially compromised the ability of the ionogels to form intimate contact with
10 surfaces, leading to poor adhesion performance of the rigid ionogels (Fig. 1c-i). When
11 the temperature exceeded T_s , the equilibrium shifted toward the disassociated state,
12 resulting in a reduction of physical crosslinking within the network and a consequent
13 decrease in mechanical stiffness. The polymer chains in the adhesive ionogels
14 underwent thermally induced disentanglement, which enhanced their mobility within
15 the gel matrix. This thermal transition facilitated the formation of softened ionogels that
16 could readily conform to the intricate contours of arbitrary surfaces (Fig. 1c-ii).

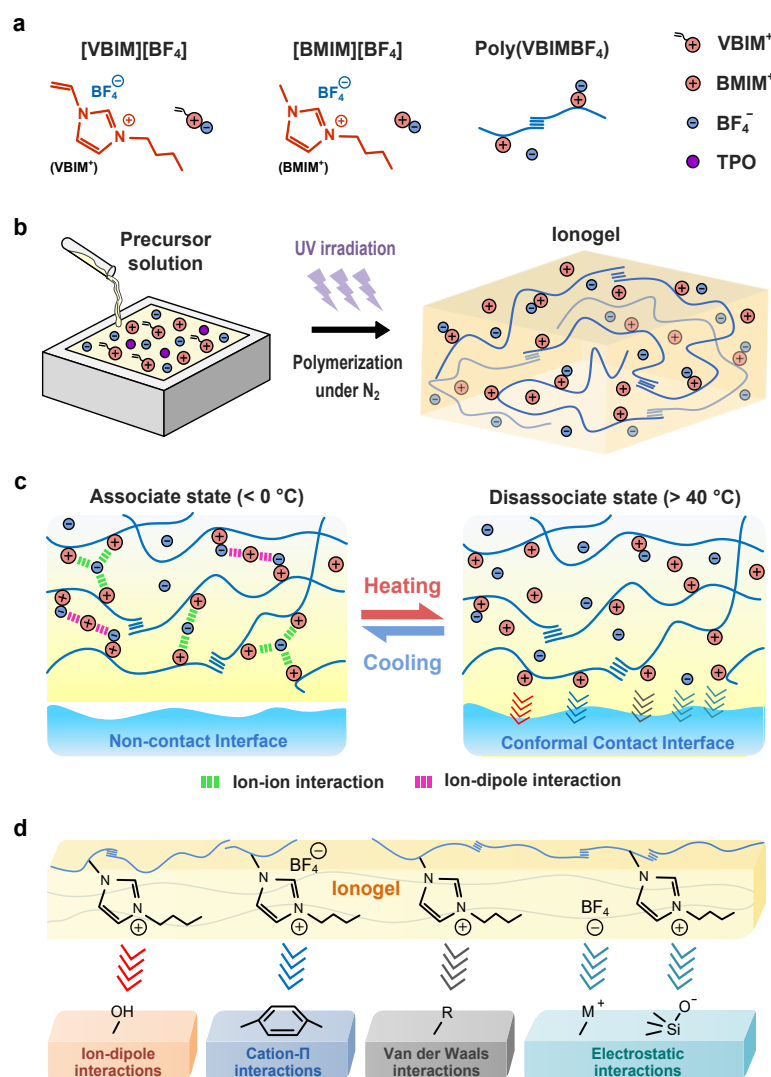


Fig. 1: Design, preparation, and working principle of PIL/IL ionogels. (a) The molecular structures of the monomers, ionic liquid, and initiator in the precursor solution. (b) A schematic illustration of the preparation process of PIL/IL ionogels. (c) The switchable adhesion mechanism of PIL/IL ionogels. (d) A schematic illustration of interfacial interactions between PIL/IL ionogels and glass.

In addition, the reconfigured polymer chains exposed an abundance of reactive functional groups, which collectively participated in the formation of multivalent interfacial bonds. These synergistic interactions led to robust and stable adhesion between the ionogels and the contacted substrates. Owing to the rich presence of functional groups within the ionogel matrix, a multitude of intermolecular

1 interactions—including ion–dipole, electrostatic, cation– π , and van der Waals forces—
2 were established at the ionogel interfaces with diverse substrates, as illustrated in Fig.
3 1d. The reversible nature of the noncovalent interactions allowed the ionogels to be
4 gently dissociated from interfaces by reducing the temperature, which highlighted their
5 promise as friendly adhesives that cause minimal damage to substrate surfaces.

6 **Characterization of Temperature-Mediated Interchain Interactions**

7 These intermolecular interactions that led to the adhesiveness established at the
8 ionogel interfaces with diverse substrates can be validated by systematically conducting
9 a series of temperature-dependent characterizations. Attenuated total reflectance
10 Fourier transform infrared (ATR-FTIR) spectroscopy was performed in the range of
11 -15°C to 60°C to evaluate the thermally induced evolution of ionic interactions within
12 the ionogels, as shown in Fig. 2a. The absorption bands observed in the range of 3200–
13 3070 cm^{-1} can be attributed to the C–H stretching ($\nu(\text{C–H})$) region of the imidazolium
14 ring in cations, while those in the range of $980\text{--}1200\text{ cm}^{-1}$ correspond to the B–F
15 stretching vibrations originating from the BF_4^- anions. With increasing temperatures,
16 the spectral intensities associated with free BF_4^- and the imidazolium groups were
17 enhanced, accompanied by a shift of the corresponding peaks toward higher
18 wavenumbers. This finding suggests that the ionic interactions among the
19 poly(VBIMBF₄) segments underwent thermal dissociation upon heating. The FTIR

1 characterization confirmed that the dynamic equilibrium of the interactions between the
2 cations, anions, and solvent molecules serves as the primary driving force behind the
3 programmable adhesion behavior of the ionogels. The mechanism underlying this
4 phenomenon is the reversible cleavage and reformation of polymer network crosslinks,
5 which regulate the exposure and shielding of functional groups.

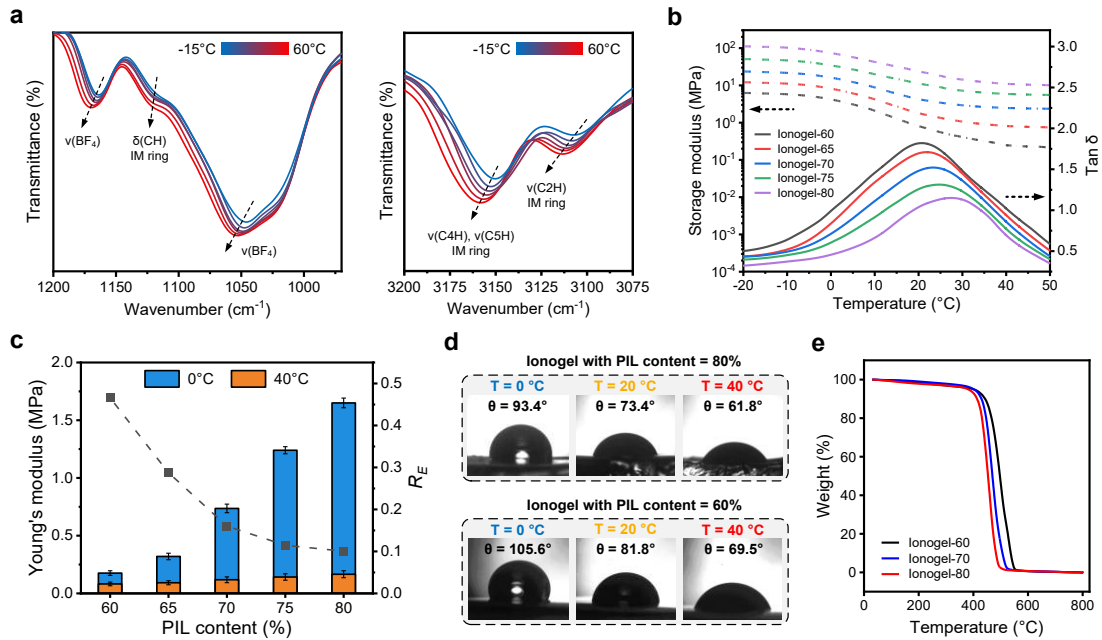
6 Rheological analyses revealed that both the storage modulus (E') and loss modulus
7 (E'') of the ionogels exhibited temperature-dependent behavior, as depicted in Fig. 2b
8 and Supplementary Fig. 4. Specifically, E' exhibited a moderate decline as the
9 temperature increased from -20°C to 50°C . This transition can be attributed to the
10 disruption of intermolecular interactions between the polymer chains. Elevated
11 temperatures led to the disassociation of these interactions, resulting in more functional
12 groups transitioning from the associated (bonded) state to the disassociated (free) state.
13 Consequently, the ionogels became increasingly soft as the temperature increased.
14 Figure 2b showed the temperature-dependent profiles of the loss factor ($\tan \delta$), with the
15 peaks of these curves determining the softening temperatures (T_s) of the ionogels. The
16 results showed that the softening temperatures of the ionogels shifted toward higher
17 temperatures as the PIL content increased. The thermal softening behavior of the
18 ionogels with different PIL contents was further confirmed via uniaxial tensile testing,
19 as shown in Fig. 2c. At 0°C (well below T_s), the ionogels became rigid and had a high

1 Young's modulus. In contrast, at 40°C (i.e., well above T_s), the ionogels became softer
2 and had a reduced Young's modulus. The modulus ratio R_E —that is, the ratio of the
3 Young's modulus at 40°C to the Young's modulus at 0°C—decreased as the PIL content
4 increased.

5 Figure 2d presented the contact angle images of water droplets on the ionogel
6 surfaces at selected temperatures for two samples with different PIL contents. At the
7 low temperature of 0°C, the ionogel substrates were intrinsically hydrophobic in nature,
8 characterized by initial contact angles close to 100°. As the temperature increased,
9 gradual reductions in the contact angles were observed, suggesting that surface
10 hydrophilicity was enhanced as the imidazolium functional groups were increasingly
11 exposed. Further, pronounced reductions in the contact angles were seen as the PIL
12 content increased (Fig. 2d), indicating the enhanced surface hydrophilicity of ionogels.
13 This trend implies that the higher PIL content level led to more exposed functional
14 groups at the interface, resulting in stronger interfacial interactions and adhesive
15 performance. The contact angle measurements of the ionogel samples with varying PIL
16 contents under different temperatures were presented in detail in Supplementary Fig. 5.

17 A thermogravimetric analysis (TGA) was conducted to evaluate the thermal
18 stability of the ionogels under elevated temperature conditions (Fig. 2e and
19 Supplementary Fig. 6). The TGA curves show that no significant weight loss was

1 detected below 350°C. The curves also indicate that the ionogels could operate across
2 a wide temperature range.



3
4 **Fig. 2: Temperature-dependent transition behavior of ionogels via interchain**
5 **interactions. (a)** Temperature-dependent FT-IR spectra of PIL/IL ionogels upon
6 heating from -15°C to 60°C (interval: 15°C). **(b)** The storage modulus (E') and loss
7 factor ($\tan \delta$) as a function of the temperature of PIL/IL ionogels. **(c)** Young's modulus
8 (E) of PIL/IL ionogels when comparing $T = 0^\circ\text{C}$ and $T = 40^\circ\text{C}$ at different monomer
9 fractions. **(d)** Evolution of the water contact angle of PIL/PL ionogels with varying
10 temperature. **(e)** TGA curves of the PIL/IL ionogels with different PIL contents.

11 Thermo-responsive adhesion performance

12 To further quantify the reversible adhesion properties of the ionogels with different
13 PIL content levels, adhesion performance was determined using three types of tests:
14 pull-off tests for pull-off adhesion strength, peeling tests for interfacial toughness, and
15 lap-shear tests for shear strength (Supplementary Fig. 7–9). Figure 3a showed the
16 results of the pull-off tests for the ionogels across a broad temperature range. The inset

1 illustrated the sample arrangement for the experiment. The tests were conducted with
2 a contact duration of 1 s and a preload of 20 kPa (Supplementary Fig. 10), in
3 accordance with standardized pull-off testing conditions. All ionogels exhibited a
4 consistent trend: The pull-off strength initially increased with temperature, reached a
5 peak, and then declined slightly. We presume that this behavior arose from a trade-off
6 between the adhesion on the contact interface and the cohesion within the ionogel
7 network. When these forces reached a balance, the ionogel could demonstrate
8 maximum adhesion strength while maintaining its shape when detached. Elevating the
9 PIL content in the ionogels from 60 wt% to 80 wt% resulted in a notable increase in
10 adhesion strength at high temperatures, from 54.7 kPa to 89.8 kPa. This improvement
11 can be attributed to two primary factors: First, increasing the PIL content enhanced the
12 internal cohesive strength of the ionogel matrix. Second, the high density of
13 imidazolium side chains on the ionogel surface facilitated stronger interfacial
14 interactions.

15 Variations in pull-off strength as a function of contact time under different
16 temperature conditions were also studied. The measured results for two temperature
17 conditions (0°C and 40°C) are shown in Fig. 3b. A rapid increase in pull-off strength
18 was observed within the first 1 s of contact under both temperature conditions. After
19 2 s, pull-off strength reached a steady value of about 75 kPa, suggesting that the

1 interfacial interactions for adhesion were established within a very short time. The
2 pull-off strength was also quantified under different preloads. Figure 3c presented
3 the development of the temperature-dependent adhesion process with preloads
4 ranging from 2 kPa to 80 kPa. A clear correlation between adhesion strength and
5 preload was observed, indicating that external mechanical loading has a noticeable
6 effect on the adhesion performance. These results suggested that both sufficient
7 contact time and increased preload effectively enhance interfacial adhesion strength,
8 likely attributable to improved surface conformity.

9 Figure 3d and Supplementary Movie 1 demonstrate the thermo-responsive
10 adhesion behavior of a square ionogel sample (10 mm side length; 2 mm thickness).
11 The ionogel was unable to adhere to the surface of a steel weight when its temperature
12 was about 0°C. However, when its temperature was higher than 25°C, the ionogel
13 enabled the immediate and robust adhesion of two weights (200 g each). The resulting
14 bonded structure exhibited remarkable mechanical stability, remaining intact under
15 substantial mechanical loads and shaking. These results indicated the tunable adhesion
16 capability of the ionogels and their potential adaptability for a broad range of practical
17 applications.

18 Figure 3e presents the pull-off adhesion strengths of the fabricated ionogel-75
19 at different temperatures across a wide range of substrates, including glass, ceramic,

1 rubber, wood, copper, paper, polydimethylsiloxane (PDMS), and various common
2 plastics, namely poly(methyl methacrylate) (PMMA), polypropylene (PP), and
3 polytetrafluoroethylene (PTFE) (Supplementary Fig. 11). The samples exhibited
4 effective adhesion at 25°C and 40°C. However, the pull-off strengths reduced by
5 approximately one order of magnitude after the samples were sufficiently cooling to
6 0 °C, allowing the ionogels to be readily detached from substrates. Specifically, for
7 a conventional glass, the pull-off strength of the ionogel decreased from 80.4 to 7.3
8 kPa. In the cases of PMMA and copper, the strengths reduced by factors of 10.6 (74.6
9 kPa to 7.06 kPa) and 9.6 (35.9 kPa to 3.75 kPa), respectively. These results
10 demonstrated the tunability of ionogel adhesiveness via temperature modulation.

11 The adhesion performance of the ionogel was quantitatively examined via 90°
12 peeling tests at 25°C. Figure 3f and 3g present the measured peeling strengths and
13 interfacial toughness of the ionogel on different substrates. These tests showed that
14 all samples exhibited interfacial failure without rupture of either the ionogel or the
15 substrates, and no ionogel residue remained on substrates after peeling. The
16 corresponding peeling strength vs. displacement curves are presented in Fig. 3f.
17 Peeling strength is defined as the peeling force divided by the width of the ionogel
18 sample. The peeling tests showed that the ionogel maintains reliable performance
19 across a range of conventional substrate materials at 25 °C, and demonstrated its

1 broadly applicable adhesive characteristics.

2 Lap-shear tests were performed for three sample strips—ionogel-75, ionogel-60,
3 and a 3M Very High Bond (VHB) tape—to study their dynamic adhesion behaviors.

4 The samples were sandwiched between two PMMA substrates (Fig. 3h) and then

5 subjected to cyclic shear deformation within a strain range of $\pm 3\%$ to generating

6 dynamic stress–strain profiles that captured their shearing adhesion performance

7 over time. The ionogel-75 strip maintained stable shear strength throughout at least

8 3000 shear cycles, whereas both the ionogel-60 and VHB tape were unable to sustain

9 the cyclic shear deformation and exhibited a significant decrease in shear strength

10 and severe failure within 1000 cycles. The pull-off adhesion strength of the ionogels

11 was further characterized over 1000 consecutive thermal cycles, consisting of

12 cooling (from 25°C to 0°C) and heating (from 0°C to 25°C) phases on a PMMA

13 substrate (Supplementary Fig. 12). As anticipated, the pull-off strength varied

14 cyclically with temperature, without notable fluctuations between each

15 heating/cooling cycle, which further confirmed the ionogels' stable and reliable

16 adhesion performance. The long-term adhesion stability of the ionogels was also

17 evaluated (Supplementary Fig. 13).

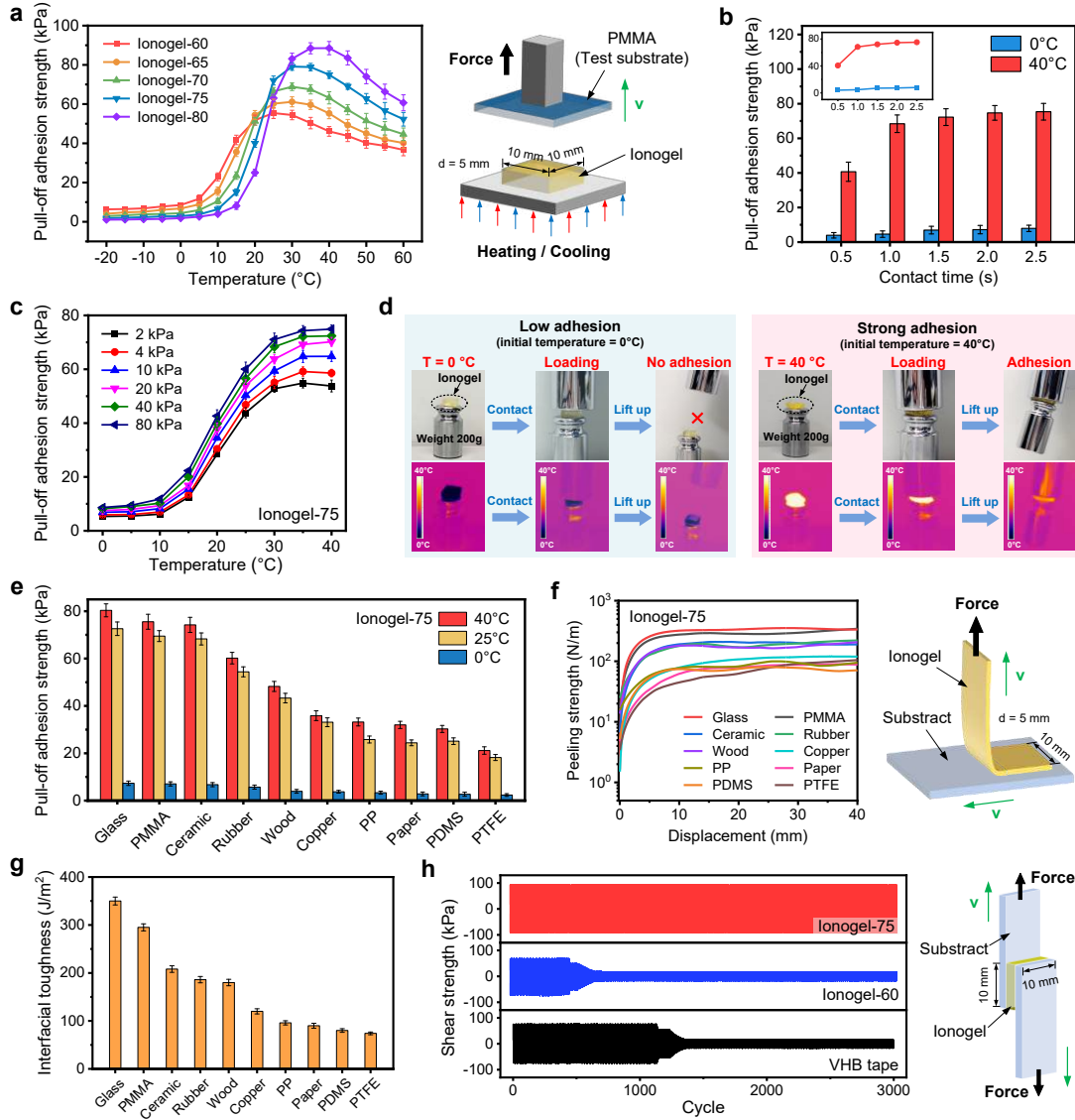


Fig. 3: Switchable adhesion properties. (a) Adhesion strength evolution of PIL/IL ionogel samples with different PIL contents versus temperature, measured through pull-off tests. The effects of (b) contact time and (c) preload on adhesion strength at different temperatures against a glass substrate. (d) The adhesion behaviors of ionogel-75 at 0°C and 40°C, respectively, show that a heavy object could be adhered and lifted easily at 40°C, but it could not be adhered at 0°C. (e) The switchable adhesion properties of ionogel-75 applied to a variety of solid substrates. (f) 90° peeling curves and (g) corresponding interfacial toughness of ionogel-75 with various substrates. (h) Lap-shear stress curves (ionogel-75, ionogel-60, and VHB tape) during repeated loading–unloading cycles until adhesion failure. The error bars correspond to the standard deviation of 3 measurements for each analysis.

1 **Potential applications**

2 The advantages of tunable adhesion capability make the ionogels produced in
3 this study well-suited to applications in climbing robots for locomotion on vertical
4 and inverted surfaces. As depicted in Fig. 4a, a tethered two-foot climbing robot
5 (Robot A) was developed for climbing demonstrations. Detailed specifications and
6 the fabrication process are provided in Supplementary Fig. 14–16. The robot
7 consisted of a pair of modular footpads connected by a double L-shaped linkage and
8 was capable of generating alternating foot motions with two motors. Each footpad
9 was assembled with an ionogel adhesion module consisting of an ionogel pad, a
10 thermoelectric plate (TEP), and a heat sink. The thermoelectric plate served as an
11 active cooling/heating device for efficient temperature regulation to modulate
12 ionogel adhesiveness (Supplementary Fig. 17).

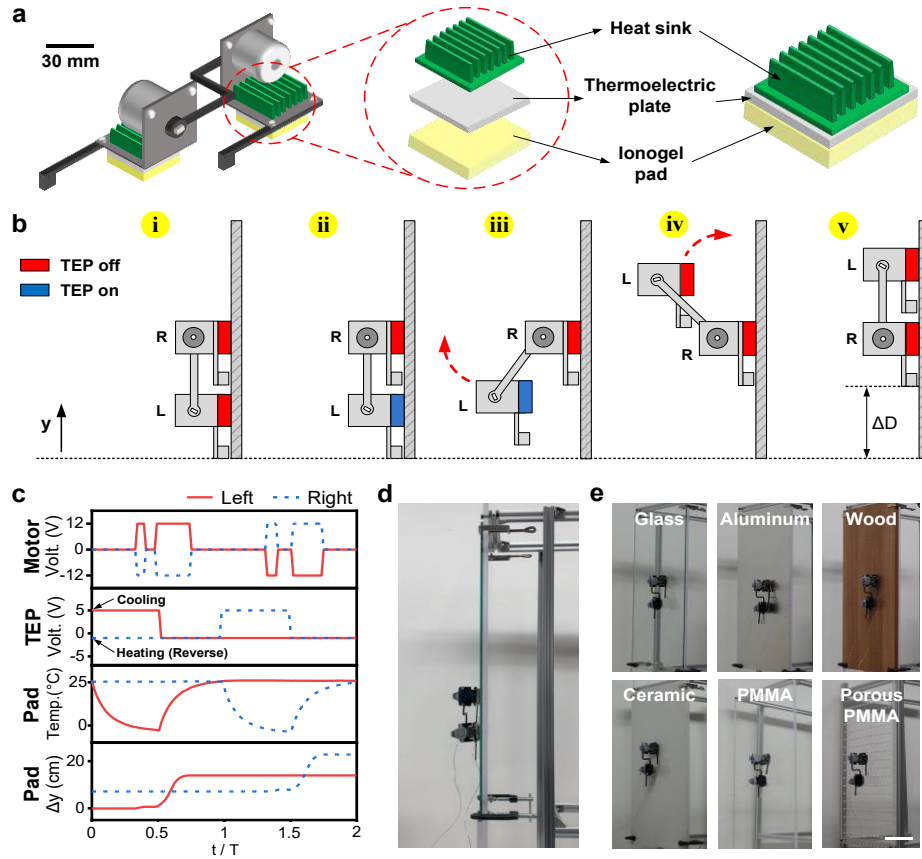
13 The wall-climbing locomotion of Robot A relied on the coordinated actions
14 between the actuation of motors and the modulation of ionogel adhesiveness on both
15 legs. Figure 4b illustrates the sequence of steps required for the robot to generate a
16 full stride over one walking cycle with a period T . Figure 4c depicts the driving
17 voltages of the motors and the TEP on both legs over two walking cycles ($2T$). The
18 measured ionogel temperature and displacement (y -direction) values are also shown
19 in the figure. Each cycle of the climbing locomotion consisted of five steps. First, the

1 robot was firmly adhered to a vertical wall, and the TEP cooling on both footpads
2 was turned off (Fig. b-i). Second, the TEP turned on, and the ionogel temperature on
3 the left footpad (the lower footpad) cooled to approximately 0°C (Fig. b-ii). Thus, its
4 adhesion significantly decreased, which allowed the left footpad to be detached from
5 the wall via motor actuation (Fig. b-iii). Upon successful detachment, the motors
6 simultaneously swung the left footpad forward until it touched the wall (Fig. b-iv).
7 Notably, a small reversed bias was applied to the TEP of the left footpad to initiate a
8 mild heating on its ionogel. Once the ionogel returned to room temperature, its
9 adhesive strength rapidly recovered, enabling the left footpad to firmly adhere to the
10 wall and complete one full climbing step (Fig. b-v). During each locomotion cycle,
11 the robot climbed up by a vertical displacement of ΔD , resulting in an average
12 climbing speed of $\Delta D/T$.

13 Figure 4d shows the experimental setup for Robot A climbing a vertical glass
14 surface with this locomotion strategy. The robot exhibited excellent climbing
15 performance on various material surfaces, including glass, a ceramic tile, an
16 aluminum plate, an acrylic plate, a wood panel, and two porous acrylic plates with
17 different pore densities (Fig. 4e, and Supplementary Movies 2–5 for six
18 representative and notably distinct substrates).

19 The vertical climbing performance of Robot A was primarily governed by the

1 shear adhesion force generated by the ionogel pad. To gain a deeper understanding
2 of its climbing capability, we measured the maximum shear adhesion force on a range
3 of surfaces (Supplementary Fig. 18). The results showed that the shear adhesion
4 forces (scaled with footpad area) significantly exceeded the weight of the robot.
5 Therefore, the adhesion capability of the ionogel pad was sufficient for supporting
6 the soft robot in maintaining stable locomotion on the material surfaces.



7
8 **Fig. 4: Design and operation principles of the two-foot climbing robot (Robot A).**
9 (a) Structure of Robot A; both feet equipped with ionogel adhesion modules. (b)
10 Schematic of the climbing principle. (c) Sequences of the control voltages for the
11 motors and the TEP modules to achieve the climbing locomotion. The measured
12 temperature and displacement are also shown. (d) Photograph of the setup for the robot
13 to climb a vertical surface. (e) Photographs of robots climbing on surfaces of different
14 materials and structures. Scale bars, 10 cm.

1 Snapshots of the wall climbing process during a locomotion cycle are presented
2 in Fig. 5a. These snapshots, labeled as states (i)–(v), are also associated with the five
3 steps illustrated in Fig. 4b. By repeating the sequential actuation steps described
4 above, long-range wall climbing was achieved. The displacements of the robot's two
5 footpads for five successive locomotion cycles are shown in Fig. 5b. The overall
6 stepwise profile reflects the robot's incremental, step-by-step locomotion. The
7 temperatures of the ionogel pads associated with the robot's movements are also
8 presented in the figure. The robot advanced approximately 73 mm per cycle, and the
9 speed of locomotion was about 58 mm min⁻¹.

10 The climbing speed of the robot was primarily influenced by the thermal
11 dissipation efficiency and pull-off adhesion performance of the ionogel pads.
12 Shortening the cooling time of the ionogel pads could effectively improve the robot
13 speed. To assess the impact of the substrate material on locomotion speed, the
14 cooling time of the ionogel pad was measured on various surfaces, including glass,
15 PMMA, wood, ceramic, CS board, and aluminum. Figure 5c presents the cooling
16 behaviors of the ionogel pad when adhering to these surfaces under an identical
17 driving voltage of the TEP. The cooling time was defined as the time required for the
18 ionogel pad to decrease from ambient temperature to 0°C. The temperature responses
19 were influenced by the substrate's thermal properties. The aluminum substrate

1 exhibited the best cooling performance because its thermal conductivity was the
2 largest among the substrates considered. The measured vertical locomotion speeds
3 across these surfaces are depicted in Fig. 5d. The results indicated that the robot
4 attained its maximum speed of 63.1 mm/min on the aluminum surface due to the
5 short cooling time for the ionogel footpad to detach during locomotion.

6 In addition to climbing vertical surfaces, the robot was capable of climbing an
7 inverted surface (i.e., a ceiling). The locomotion principle of climbing inverted
8 surfaces is similar to that of vertical surfaces, except that a longer ionogel heating
9 time is required to enhance ionogel adhesiveness for inverted surface climbing. This
10 is because the upward normal adhesion force of ionogel plays a crucial role in
11 counteracting gravitational force along the direction perpendicular to the inverted
12 surface. Supplementary Fig. 19 illustrates the experimental setup for testing the
13 robot's movement along an inverted surface. A sequence of snapshots taken over a
14 locomotion cycle is shown in Fig. 6e (Supplementary Movie 6). The average
15 locomotion speed was approximately 56 mm/min.

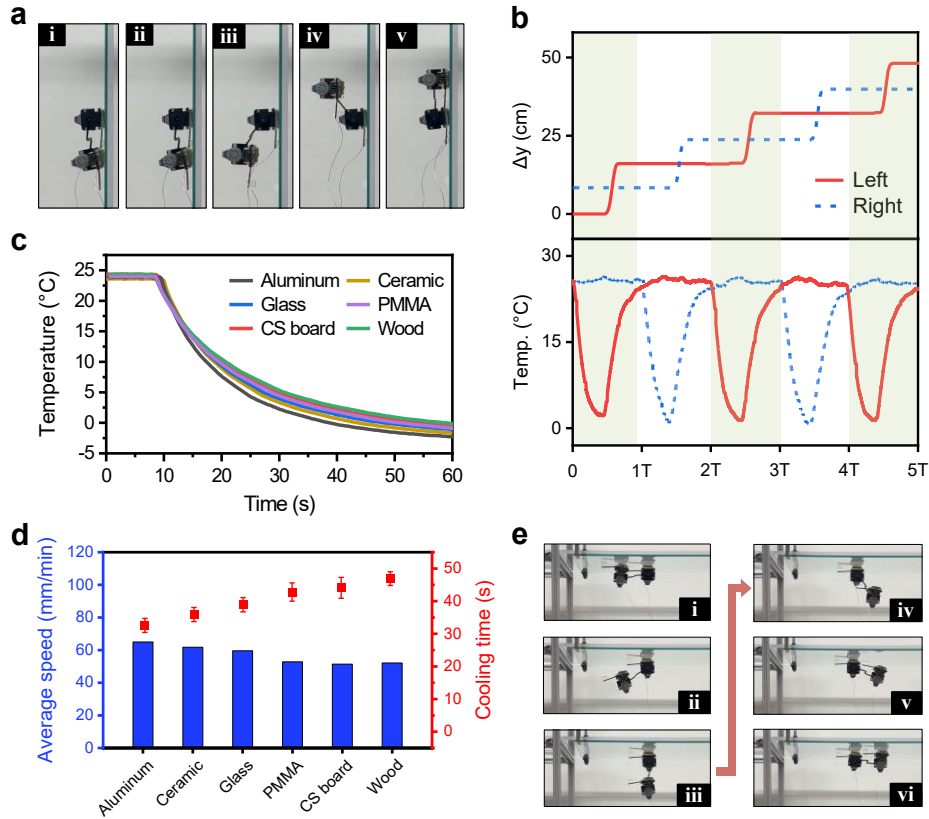


Fig. 5: Climbing performances of the two-foot climbing robot (Robot A) on vertical and inverted surfaces. (a) Snapshots of wall climbing on a glass surface during a locomotion cycle; and **(b)** the associated vertical displacement and temperature vs. time. **(c)** Cooling curves of a PIL/IL ionogel sample on different surfaces. **(d)** Locomotion speeds of the robot when climbing on different substrate materials. The average cooling times required for detaching the PIL/IL ionogel pads from each substrate are also given in the figure. **(e)** Snapshots of inverted climbing on a glass surface.

Furthermore, we designed a three-foot climbing robot (Robot B) for demonstrating load carrying. The schematic is shown in Fig. 6a. Detailed information regarding Robot B is provided in Supplementary Fig. 20. The ionogel pads mounted on each foot were approximately $14 \text{ mm} \times 14 \text{ mm} \times 2 \text{ mm}$. The weight of the robot is approximately 98 g. A load-carrying platform or fixture can be easily assembled on the middle foot of Robot B. Figure 6b–6c and Supplementary movies 7–8 demonstrate that Robot B

1 climbed a vertical PMMA substrate while carrying different payloads (i.e., 100, and
2 150 g). The maximum load-bearing capacity is approximately 19.1 g/cm² during
3 locomotion on a vertical wall. Notably, the robot was able to support its own weight
4 of 98 g while carrying an additional payload of 150 g, which is about 153% of its
5 own weight. The demonstrated load-carrying capability not only indicates that the robot
6 possesses sufficient capacity to carry onboard batteries, electronics and payload, but
7 also facilitates the applications requiring autonomous untethered climbing motion on
8 vertical surfaces with cargo transportation.

9 We also leveraged the reversible adhesion properties of the PIL/IL ionogels to
10 develop a small vertical transport tower capable of vertically transporting a cargo by
11 repeatedly capturing, transporting, and releasing objects. Figure 6d illustrated the
12 simplest version of the vertical transport tower which consists of two transferring
13 pads. Each transferring pad comprised an ionogel adhesion module, a servo motor,
14 and a short arm. Figure 6e and Supplementary Fig. 21 depict the process of vertically
15 conveying an object from a lower transferring pad to an upper transferring pad. The
16 transport tower presented here can be readily expanded by integrating multiple
17 modules to achieve scalable and flexible transport operations (Supplementary Movie
18 9).

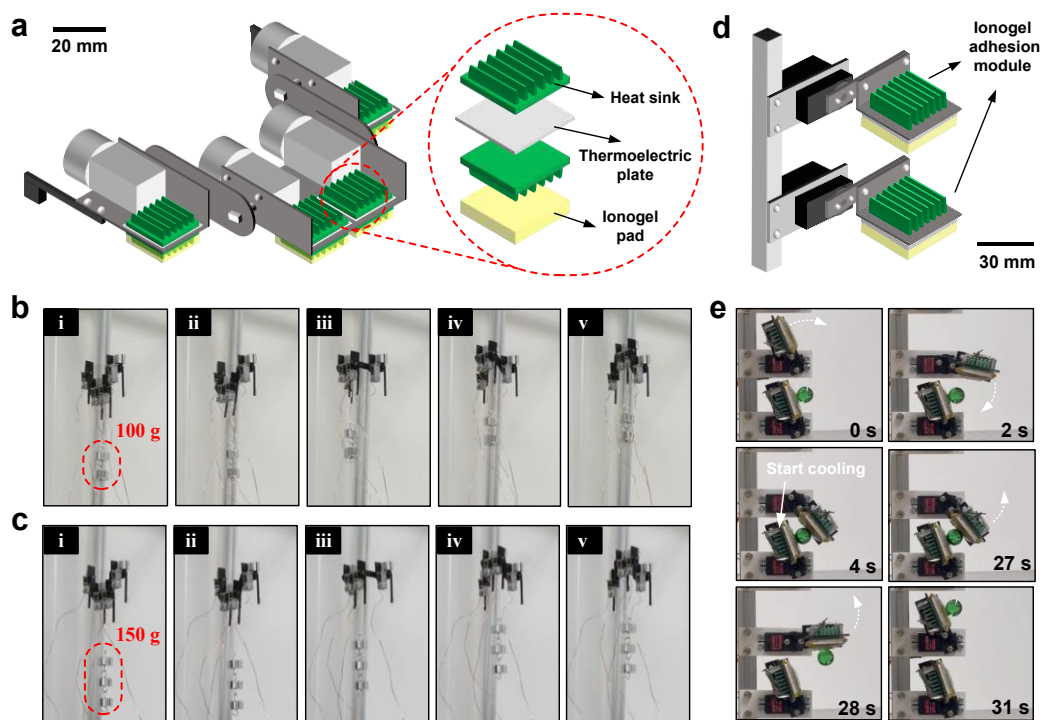


Fig. 6: Demonstrations of vertical load transportation with switchable adhesive ionogel. (a) Schematic of a three-foot climbing robot (Robot B). Snapshots of the climbing process with a (b) 100-g; (c) 150-g payload on the PMMA substrate during a locomotion cycle. (d) Schematic of a vertical load transportation tower consisting of two ionogel adhesion modules. (e) Sequential snapshots of picking up and releasing a glass marble by the transportation tower.

In summary, the PIL/IL ionogels exhibited pronounced adhesion switchability in response to temperature modulation, which was enabled by reversible dynamic physical cross-linking within the ionogel network. By varying the identities of the constituent cations and anions, the ionogels achieved a favorable balance between internal cohesive forces and ionogel–substrate interfacial interactions. By attaching the ionogels to the moving parts of simple two-foot and three-foot robots, we achieved climbing locomotion on vertical and inverted walls. This study demonstrates that longstanding challenges in robotic climbing—such as the reliance on complex

1 materials and microstructures on contact interfaces, intricate device designs, and high
2 engineering thresholds—can potentially be addressed through a polymer network
3 regulation strategy. We anticipate that elucidating the adhesive mechanisms governed
4 by dynamic interchain interactions can facilitate the practical design of soft adhesive
5 materials with advanced, unconventional functionalities. Although we primarily
6 focused on PIL/IL-based ionogels that exhibit characteristics of temperature-induced
7 variations in adhesion strength, modulus, and wettability, the strategy and
8 mechanisms proposed herein may be extendable to the development of other smart
9 soft materials through the coupling of multiscale interchain interactions, thereby
10 broadening the application scope of smart materials.

11 **Methods**

12 **Materials**

13 Ionic liquid monomer 1-vinyl-3-butyylimidazolium tetrafluoroborate (VBIMBF₄,
14 98%), ionic liquid 1-butyl-3-methyylimidazolium tetrafluoroborate (BMIMBF₄, 99%),
15 and ethyl (2,4,6-trimethylbenzoyl) phenylphosphinate (TPO-L, 95%; photoinitiator)
16 were obtained from Echo Chemical Co., Ltd. All chemicals were used as received
17 without further purification. Prior to use, the ionic liquids were dried under vacuum
18 at 80°C for 48 h and purged with N₂ to ensure an oxygen-free environment.

1 **Preparation of PIL/IL ionogels**

2 PIL/IL ionogels were fabricated via a one-step, photoinitiated homogeneous
3 polymerization process in the absence of cross-linkers. All preparation procedures
4 were conducted in a N₂-filled glovebox. Specifically, 1.2 g of VBIMBF₄ monomer
5 and 12 mg of ethyl (2,4,6-trimethylbenzoyl) phenylphosphinate (1.0 wt% relative to
6 the monomer) were completely dissolved in 0.8 g of BMIMBF₄ at 25°C. The
7 resulting solution was ultrasonicated for 30 min, degassed under vacuum conditions
8 for 30 min, and subsequently allowed to stand undisturbed for 2 h prior to
9 polymerization. The precursor solution was then transferred into a custom-made
10 poly(methyl methacrylate) (PMMA) mold and irradiated with UV light (8 W, 365
11 nm) for 24 h to obtain ionogel-60. Other ionogels were prepared using the same
12 procedure, and their detailed compositions are listed in Supplementary Table S1.

13 **Characterization and Instruments**

14 **Rheological test.** The dynamic mechanical properties of the ionogels (30 mm length,
15 5 mm width, and 2 mm thickness) were characterized using a dynamic mechanical
16 analyzer (DMA Q800, TA Instruments, USA) in temperature-ramp mode. Each
17 sample was cooled to -20°C for 3 min prior to testing. The measurements of
18 temperature sweeps, which was ramped from -20 to 50 °C at 2 °C min⁻¹, were
19 performed in submersion tensile mode at 0.1% strain and 1 Hz with a 0.1 N preload.

1 **Mechanical properties test.** The temperature-dependent tensile properties of the
2 ionogels were measured using a dynamic mechanical analyzer (DMA Q800, TA
3 Instruments, USA). Dumbbell-shaped specimens (30 mm length, 5 mm width; 3 mm
4 inner width, 10 mm gauge length, and 2 mm thickness) were tested at a deformation
5 rate of 5 mm min⁻¹. The specimens were elongated to failure, and stress–strain curves
6 were recorded. Young’s modulus (E) was determined from the initial linear region
7 as the slope between 5–15% strain. To obtain stress–strain responses at high and low
8 temperatures, tests were performed at specified temperatures. For each measurement
9 condition, at least three replicates were measured, and averages were reported.

10 **Fourier transform infrared spectroscopy test.** FTIR spectroscopy with attenuated
11 total reflectance (ATR) mode (Spectrum Two, PerkinElmer, USA) was used to
12 characterize the ionogel samples. Spectra were acquired over 400–4000 cm⁻¹ with 12
13 co-added scans at a spectral resolution of 1 cm⁻¹. The samples were heated from -
14 15°C to 60°C at 15 °C increments to obtain temperature-controlled measurements.
15 Each setpoint of temperature was held for 10 min to reach equilibrium. To investigate
16 the ionogel formation process, the FTIR spectra of the precursor solution were
17 recorded after 0 and 24 h of UV irradiation.

18 **Contact angle test.** Static water contact angle measurements were taken to determine
19 the hydrophilicity of the ionogel samples at different temperatures. the measurement,

1 the ionogel sample was fixed at a hot plate for the cases of 20 °C and 40 °C, and fixed
2 at a thermoelectric cooling plate for the case of 0 °C. A contact angle goniometer
3 (FTA125, First Ten Angstroms, USA) was used to perform static water contact angle
4 measurements. Contact angle values were recorded and averaged for each condition.

5 **Adhesion performance measurement.**

6 Adhesion tests were performed using a XYZ positioning stage (SR-300C, LTE,
7 Taiwan) equipped with a digital force gauge (HF-1, Algol, Taiwan) (Supplementary
8 Fig. S7–S9) to quantify the interfacial strengths of ionogel samples on various
9 substrates.

10 For pull-off tests, temperature was controlled by adjusting either a heating plate or a
11 thermoelectric cooling plate placed beneath the substrate, which enabled the
12 measurement of adhesive strength at various temperatures. Temperatures were
13 verified using an infrared camera. To measure their pull-off strengths, the ionogels
14 were cut into samples of 10 mm × 10 mm × 5 mm, placed between two identical
15 substrates, compressed with a 2 N preload for 1 s, and then separated at a constant
16 crosshead speed of 50 mm min⁻¹. Prior to testing, all substrates were cleaned with
17 ethanol and water and dried. Applied force was obtained from the adhesion (force–
18 time) curve, and pull-off strength was calculated as the maximum force divided by
19 the bonded area. For each condition, three replicate measurements were obtained,

1 and the average pull-off strength was reported.

2 To measure interfacial toughness, adhered samples (10 mm in width) were subjected

3 to 90° peel tests. A nonextensible tape was used as a backing layer to prevent

4 elongation of the ionogels during testing. The peel rate was maintained at 50

5 mm/min⁻¹. A steady-state peeling force was observed, as the force plateaued during

6 the test. Interfacial toughness was calculated as the ratio of the plateau force to the

7 sample width.

8 For shear strength calculations, lap-shear tests were performed on adhered samples

9 with a bond area of 1 cm × 1 cm. The peel rate for this test was maintained at 50

10 mm/min⁻¹. Shear strength was calculated as the maximum applied force divided by

11 the bonded area.

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- 1 experimental results. Y.-J.Y. and Y.-C.C. conceived the idea, designed the research,
- 2 and co-wrote the manuscript. Y.-J.Y. supervised the project.

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- 5 **Competing interests**

- 6 The authors declare no competing interests.

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