

This is an Open Access document downloaded from ORCA, Cardiff University's institutional repository: <https://orca.cardiff.ac.uk/id/eprint/167894/>

This is the author's version of a work that was submitted to / accepted for publication.

Citation for final published version:

Lu, Shan, Ma, Zeyu, Ding, Mingwei, Wu, Yuhao, Chen, Yuanli, Dong, Mochen and Qin, Liguang 2024. Reversible electroadhesion induced through low ion concentration migration for biomedical applications. Chemical Engineering Journal 486 , 150393. 10.1016/j.cej.2024.150393

Publishers page: <http://dx.doi.org/10.1016/j.cej.2024.150393>

Please note:

Changes made as a result of publishing processes such as copy-editing, formatting and page numbers may not be reflected in this version. For the definitive version of this publication, please refer to the published source. You are advised to consult the publisher's version if you wish to cite this paper.

This version is being made available in accordance with publisher policies. See <http://orca.cf.ac.uk/policies.html> for usage policies. Copyright and moral rights for publications made available in ORCA are retained by the copyright holders.



Reversible electroadhesion induced through low ion concentration migration for biomedical applications

Shan Lu ^a, Zeyu Ma ^a, Mingwei Ding ^a, Yuhao Wu ^a, Yuanli Chen ^b, Mochen Dong ^c, Liguo Qin ^{a,*}

^a Key Laboratory of Education Ministry for Modern Design and Rotor-Bearing System, Institute of Design Science and Basic Components, Xi'an Jiaotong University, Xi'an 710049, China

^b State Key Laboratory of Military Stomatology & National Clinical Research Center for Oral Diseases & Shaanxi Clinical Research Center for Oral Diseases, Department of Craniofacial Trauma and Orthognathic Surgery, School of Stomatology, FMMU, Xi'an, China

^c School of Chemistry, Cardiff University, Cardiff CF10 3AT, United Kingdom

ARTICLE INFO

Keywords:

Reversible electroadhesion
Carrageenan
Ion migration
Hydrogel

ABSTRACT

Reversible adhesions have exhibited a great potential in the biomedical field. Here, we designed a reversible electroadhesion between anionic κ-carrageenan hydrogel and physiological saline-based nonionic polyacrylamide hydrogel through the low and controllable ion migration. A robust adhesion was obtained between the hydrogel pairs via a direct current (DC) of 10 V. Upon the application of a reverse voltage, the pair could be effortlessly and rapidly detached. We elucidated the mechanism of reversible electroadhesion through electrochemical analysis, attributing the adhesion to the formation of double ion layers between the adhesive interfaces under the electrical field and pH variation. When a reverse voltage was applied, the adhesion strength was dropped lower than 0.1 kPa and the detachment was occurred due to the disruption of the double ion layer by ionic migration and even diffusion. Moreover, the adhesion system demonstrated outstanding electrochemical stability, with less than a 7 % variation in charge storage capability after 1000 cycles. The proposed strategy paves the way for developing high-performance adhesion hydrogels with electrically controllable functions, which are indispensable for various emerging fields like tissue repair and soft biomedical applications.

1. Introduction

Reversible electroadhesion is a fascinating phenomenon where the application of a forward voltage induces adhesion between two components, while a reverse voltage facilitates their separation [1]. This unique property allows for rapid and controlled attachment and detachment, making it particularly useful in various applications. For instance, in biomedical scenarios, reversible adhesion can be employed for temporary attachment to human tissues during medical procedures, providing both precision and safety [2–5]. Similarly, in the field of robotics, this technology enables climbing robots to adhere to and detach from walls and other surfaces effortlessly, enhancing their mobility and utility in complex environments [6]. The versatility and efficiency of reversible electroadhesion have thus sparked considerable interest and innovation across both biomedicine and robotics, promising a range of groundbreaking applications and advancements.

Typically, the process of achieving electroadhesion between interfaces necessitates the application of high voltages, often in the

kilovolt (kV) range [7]. These high voltages are required to break down the barrier at the interface and establish an electrostatic layer, which facilitates adhesion. However, a significant drawback of using such high voltages is the risk of causing irreversible damage to the materials involved. This challenge limits the practicality and safety of traditional electroadhesion methods, particularly in delicate applications like biomedicine or sophisticated robotics. In contrast, there have been intriguing reports of successful electro-adhesion between gel materials using much lower voltages [8–10]. This advancement is noteworthy because it suggests the possibility of achieving effective electroadhesion without the associated risks of high-voltage applications. However, the mechanisms underlying this low-voltage electro-adhesion in gels are not yet fully understood. A deeper exploration into these mechanisms is crucial for developing more efficient, safer, and versatile electro-adhesion technologies. Such understanding could potentially lead to broader applications and innovations in fields where gentle yet firm adhesion is required, such as in wearable medical devices or in robotics where delicate handling is essential.

* Corresponding author.

E-mail address: liguoqin@mail.xjtu.edu.cn (L. Qin).

Typical adhesion is commonly caused by mechanisms such as the covalent cross-linking between materials, electrostatic interactions, hydrogen bonding, and atmospheric pressure [6,11]. These mechanisms result in adhesion that is difficult to separate once formed, and even more challenging to achieve controlled reversible adhesion. In this study, we introduce a reversible adhesion controlled by positive and negative voltages, enabling controlled adhesion between hydrogels in a particularly simple manner. By using physiological saline to prepare polyacrylamide (PAAm) hydrogels, we created a medium that closely simulates the characteristics of real human tissues. The key to our approach lies in the use of κ -carrageenan (KC), a substance known for its molecular chains that carry a negative charge [12,13]. The interface between the two achieved reversible adhesion under low voltage conditions. Subsequently, we employed electrochemical methods to elucidate the mechanism behind the formation of reversible adhesion, tested the voltages required for creating and releasing adhesion, and verified the stability of the cyclic reversibility. Our studies collectively demonstrate the potential utility of electroadhesion in biomedical applications.

2. Experimental procedure

2.1. Materials

κ -carrageenan (KC) was provided by Solarbio. The monomer acrylamide (AAM), crosslinker N,N'-methylenebisacrylamide (MBAA), initiator 2-Hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone sodium chloride (NaCl) and potassium chloride (KCl) were purchased from Aladdin. All material were used as received. Deionized (DI) water was used in all experiments.

2.2. Synthesis of κ -carrageenan hydrogels

Anionic KC hydrogels were prepared using the following protocol. 0.2 g of KC, 0.002 g KCl and 10 mL DI water were magnetic vortexed at 60 °C water bath condition for 10 min. In order to remove dissolved gas, the solution was sonicated by ultrasonic crusher for 3 min. Molds were made by silicone rubber, and glass plates as the cover, respectively. The solution was poured onto the mold (4 cm × 6 cm × 1 mm) and sealed with the cover, cooled to room temperature and cured naturally.

2.3. Synthesis of PAAm hydrogels

PAAm hydrogels were prepared by a variation of the method described by Kim et al [14]. First, preparing 0.9 wt% physiological saline solution. Next, 5 g AAM, 0.005 g MBAA, 0.004 g initiator were added into 10 mL physiological saline solution. The mixture was vortexed for 1 min. Molds were made by glass, poured the solution onto the molds and sealed with the cover. UV light (365 nm, 20W) was irradiated for 1.5 h for curve.

2.4. Adhesive experiments

To measure interfacial toughness, adhered hydrogels with 10 × 20 mm² were prepared, fixed on the upper and lower fixtures of the tensile tester (ZQ990, ZhiQu Precision Indus, China) and control the contact area of two hydrogels to 10 × 10 mm². The lap-shear test was used for adhesive toughness (ASTM F2255) at 20 °C and relative humidity of 40% – 60%. A DC power source (A-BF, model SS-305S, 0–30 V, 0–5 A) and graphite electrodes (from Tianwang, China) were connected to the DC power and apply voltage to the hydrogel – hydrogel pair. Different voltages were applied from +10 V to –10 V for 10 s. After energizing, the two gels were stretched at a speed of 5 mm/min until separation and the stress–strain curve was recorded. All experiments were repeated at least three times. The surface morphology after adhesion was observed by a confocal laser scanning microscope (CLSM, Leica DCM, Germany).

For hydrogel–tissue tests in Table 2, we employed a subjective

evaluation method by two observers to assess the adhesion strength [10]. The porcine tissues used were provided by the Air Force Medical University, at least three tissue samples and three KC hydrogels were prepared. First, we detected the direct adhesion between the hydrogel and the tissue, and then the adhesion strength between them after electrification. The adhesion strength is categorized into five levels 0–4, where 0 = negligible, 1 = weak, 2 = moderate, 3 = strong, and 4 = very strong adhesion. Two observers independently evaluated the adhesion strength, and the average of their evaluations was taken as the data in the Table 2.

2.5. Electrochemistry characterizations

Electrochemical measurements were measured by using an electrochemical workstation (ChenHua, CHI660E, China) with Ag/AgCl electrode, Pt plate and adhesive specimen as reference, contour and working electrodes constituted a three-electrode system in physiological saline solution. Cyclic voltammetry (CV) tests from –3 V to +3 V to –3 V at 100 mV/s was tested for 1 cycle, and from –1.5 V to +1.5 V to –1.5 V at 500 mV/s was tested for 1000 cycles. The Charge storage capability (CSC) of the sample was calculated from the measured CV results as:

$$CSC = \frac{\int_{E_1}^{E_2} i(E) dE}{E_2 - E_1 \cdot 2vA}$$

Where E_1 and E_2 are the potential window, i is the current, v is the scan rate, and A is the area of the adhesive contact area, respectively.

Electrochemical impedance spectroscopy (EIS) was carried out with the frequency range of 10⁵ to 10⁻¹ Hz and voltage was based on open circuit voltage from +2 V to –0.75 V. Experimental EIS data were fitted using ZView software.

3. Results and discussion

3.1. Reversible adhesion strength

Initially, the adhesion strength between interfaces was evaluated using KC hydrogels and PAAm hydrogels under bias voltages (± 10 V). A tensile tester was employed to assess the shear strength between the two hydrogels [10,15] (as Fig. 1 schematic shows), applying stress to the bonding surface until failure occurred. The stress–strain curves of the adherent interface at zero voltage and under bias voltage conditions are presented in Fig. 1A. Here, zero voltage denotes the measurements taken with the unenergized sample when the two materials are pressed together, while the positive voltage signifies the PAAm hydrogel connected to the positive terminal and the KC hydrogel to the negative terminal. The stress–strain curves for forward bias, as shown in Fig. 1A, extended to higher stresses than those observed for contact adhesion, indicative of the enhanced adhesion strength provided by the electric field. Conversely, the adhesion strength significantly diminished when a reverse bias was applied. The adhesion shear strength between the two hydrogels was determined with the following equation:

$$\text{Shear strength} = \frac{F_{max}}{WL}$$

where F_{max} is the maximum force in lap-shear and tensile test, W is width, L is length.

As Fig. 1B shows, at zero voltage, the shear adhesion strength measures approximately 3.35 kPa, whereas under forward bias, it nearly doubles, reaching around 6.2 kPa. Upon testing the failed gel under forward voltage bias, it was observed that failure typically results from the breaking of KC, suggesting that the adhesion strength could potentially achieve even higher levels [10]. Under reverse bias, the adhesion strength was nearly zero. In practical tests, applying reverse bias typically results in the immediate separation of the interface between the two materials, leading to a complete loss of adhesion performance

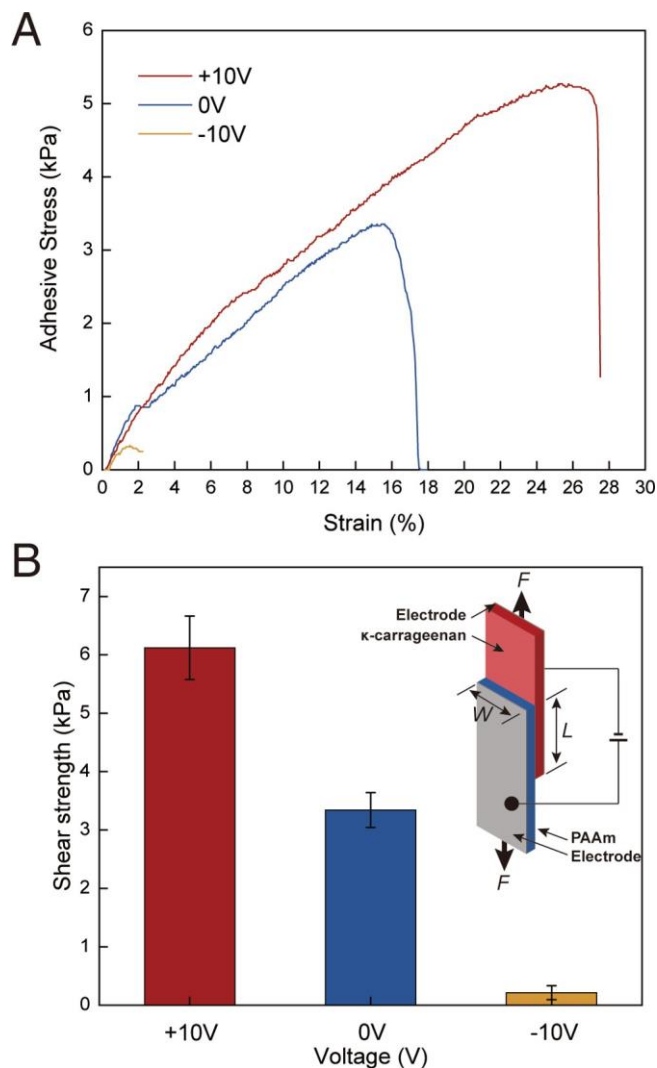


Fig. 1. (A) Stress-strain curves of shear adhesion under varying voltages. (B) Shear strength calculated at different voltages.

(Movie S1).

Here, we demonstrate the reversible adhesion properties between hydrogels, exhibiting nearly a change of 20 times in adhesion strength under different bias voltages. Lap-shear tests also revealed distinct fracture elongations associated with adhesive failure, attributed to the progressive increase in adhesive force during adhesion. Initially, the hydrogels utilize stretching to overcome the adhesive force, until reaching the shear strength limit where interfacial sliding occurred and adhesive failure was happened. Comparing with typical adhesion mechanisms such as interfacial melting or physical entanglement, the destruction of the adhesive interface typically results in surface morphology damage, preventing secondary adhesion. In contrast, the reversible adhesion we demonstrate allows for numerous cycles of reversible adhesion without exhibiting melting phenomena. Additionally, utilizing confocal laser microscopy (CLSM), we captured the interface morphology after ten cycles of adhesion, which remained remarkably smooth, indicating that the adhesion is not a result of melting or physical entanglement phenomena (Fig. S1).

3.2. Reversible adhesion mechanism

Due to their high water content, hydrogels are examined using electrochemical impedance spectroscopy (EIS) to analyse the reversible adhesion phenomenon occurring during the energization process

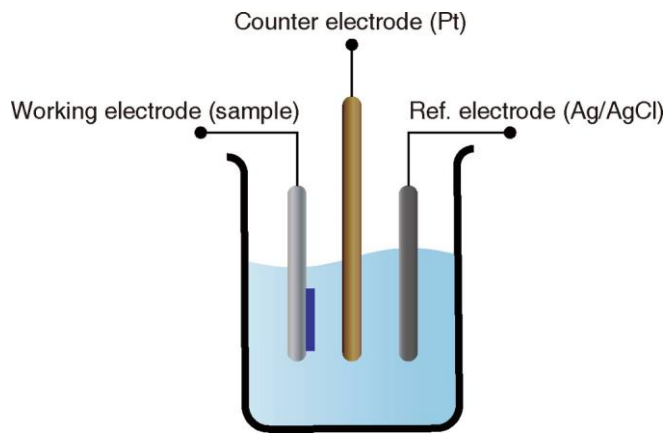


Fig. 2. Schematic illustration of an electrochemistry setup for adhesion layer.

(Fig. 2). The overall open-circuit voltage was firstly measured, followed by the application of bias voltages with the open-circuit voltage as the reference zero potential.

The EIS results of the adhesion interface at zero voltage were initially examined, as depicted in Fig. 3. Here we design that when voltage is applied, an adhesion effect is achieved by the formation of a capacitive

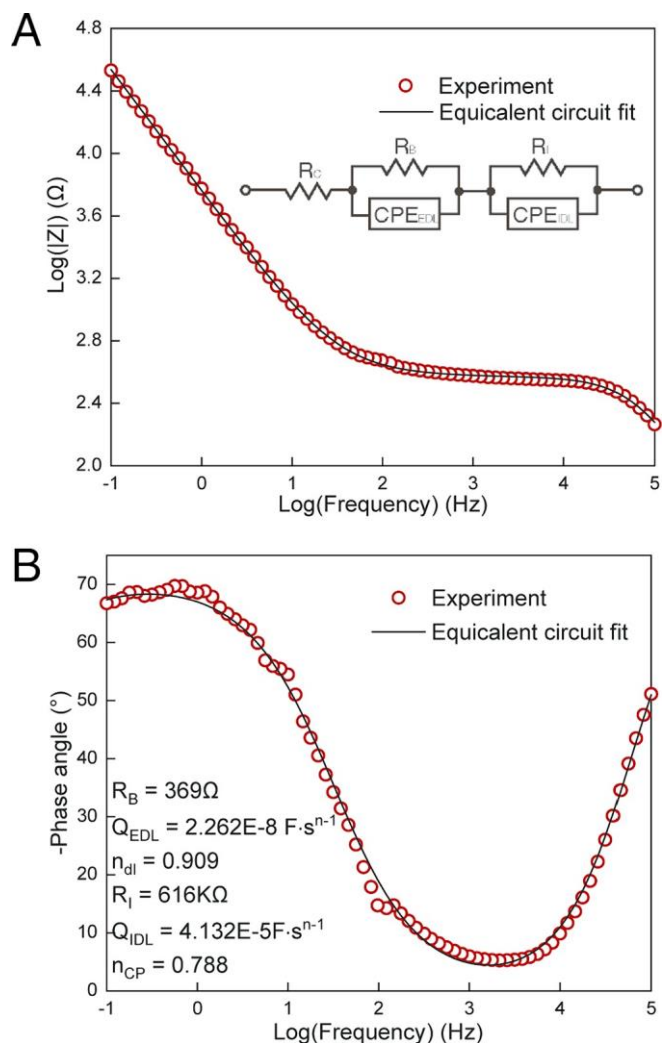


Fig. 3. (A) Impedance Bode plot and fitting circuit at open circuit voltage. (B) Bode phase plots.

layer at the interface, due the Na^+ ions in the PAAm migrate to the interface, the negatively charged carrageenan molecular chains moving towards the vicinity of the interface, and this simulated circuit demonstrates exceptional fitting accuracy. This model contains the Ion Double Layer (IDL) at the interface CPE_{IDL} is attributed to ion migration, the interfacial ionic current R_i , the resistance of the drift of free ions R_B , the electrical double layer (EDL) at the electrode/hydrogel interface CPE_{EDL} , the contact resistance R_C . Based on this assumption, a simulated circuit was utilized to fit the data and explain the causes of adhesion, as illustrated in Fig. 4A. In this simulated circuit, as the voltage increases, the capacitance may gradually experiences breakdown, resulting in a progressive decrease in R_i [1]. When all other parameters are held constant and only the R_i value is altered for fitting, the resulting curve is as depicted in Fig. S2. The fitted results indicate a gradual decrease in impedance values ($|Z|$) at low frequency, and the phase angle progressively approaches zero.

To validate the accuracy of this fitting, various bias voltages (from 2 V to -0.75 V) were applied using the open-circuit voltage as a baseline, and the EIS spectra of the adhesion interface were tested. When a positive bias voltage is applied, with an increase in voltage, the impedance values at low frequencies significantly decrease (Fig. 5A). This is accompanied by the phase angle approaching zero at low frequencies (Fig. 5B). The Nyquist plot maintains a semi-circle shape, and fitting using the above circuit shows the interfacial resistance (R_i) (Table 1) decreases significantly with voltage increase also demonstrates excellent results. When reverse bias voltage was applied, no significant decrement of the impedance ($|Z|$) was observed, however, a noticeable reduction at low frequency was noted in the phase angle spectra, which means that the circuit fitting results with the circuit shown in Fig. 4A were not accurate under reversed bias voltage (Fig. 5C), suggesting the possibility of a novel electrochemical phenomenon. We found that the Nyquist plot (Fig. 5C) deviated from a circular shape, exhibiting an increase in impedance values at low frequencies, a phenomenon attributable to ion diffusion under reverse bias. Under higher voltage, ions in aqueous solution media migrate towards the electrodes, even crossing interfaces, resulting in the diffusion phenomena observed in the EIS results (-0.75 V).

As Na^+ ions originating from PAAm and KC chains and migrating towards the interface under a forward bias, they establish an ionic

double layer which facilitate electrostatic interactions and subsequent adhesion. With the gradual increase in voltage, the capacitance layer is progressively penetrated, reducing the impedance of the interfacial double layer (IDL), thus augmenting the adhesive force at the interface. Conversely, under reverse bias conditions, Cl^- ions from PAAm and K^+ ions from KC migrate towards the interface, readily forming ion pairs within the liquid medium. This perspective is further supported by the observed electrochemical diffusion phenomenon (Fig. 5C, yellow curve) upon further voltage increase. To clearly verify the diffusion of ions in KC, we connected KC with Q235 steel and applied electricity. Remarkably, the electrolytically generated Fe^{3+} ions migrated into the KC solution, resulting in a distinctive brownish-yellow coloration (Fig. S3). This migration of iron ions, as described, has been previously researched [16] and similarly was regarded as the robust adhesive forces.

To more accurately track the electrochemical process during the adhesiveness variation, we acquired the CV curve using a three-electrode system (potential window, -3.0 to 3.0 V). The corresponding CV curve (Fig. 6) shows that the onset potentials of oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) are 1.50 V and -0.88 V (versus Ag/AgCl), respectively. The electrochemical analyses elucidated a pivotal aspect concerning the electrically reversible adhesion of hydrogels, indicating that the activation of adhesiveness necessitates voltages surpassing the onset potential of the OER [17,18]. To corroborate this perspective, we detected pH variations at the interface. Upon applying positive bias voltage, the pH at the KC interface was transitioned from 7 to 13. While the reverse bias voltage was applied, the pH value was shifted from 13 to 3. The pH variations observed at the interface offer additional evidence supporting the role of electrochemical reactions in adhesion modulation. The transition from neutral pH to alkaline pH upon application of positive bias voltage indicates the generation of hydroxide ions, likely through the OER. Conversely, the decrease in pH upon reverse bias voltage suggests the occurrence of hydrolysis reactions, possibly associated with the reduction in pH observed during HER [19]. These pH variations corroborated the involvement of electrochemical processes, in driving adhesion transitions in hydrogels.

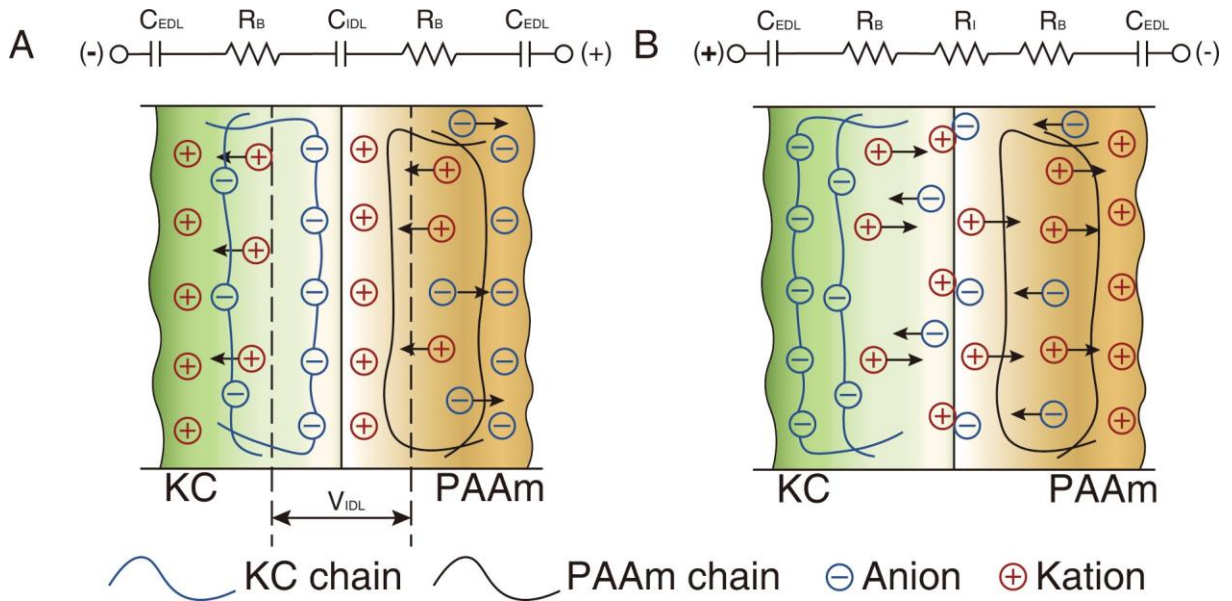


Fig. 4. Schematic illustration of hydrogel adhesion operating under forward bias voltage (A) and reverse bias voltage (B). (A) Under forward bias, KC is connected to the negative terminal of the power source, and PAAm to the positive terminal, with the voltage drop across the Interfacial Double Layer (IDL) providing electrostatic adhesion. (B) Under reverse bias, the kation ions from KC and Anion from PAAm meet at the interface, disrupting the electric field within the IDL, significantly reducing the adhesive force.

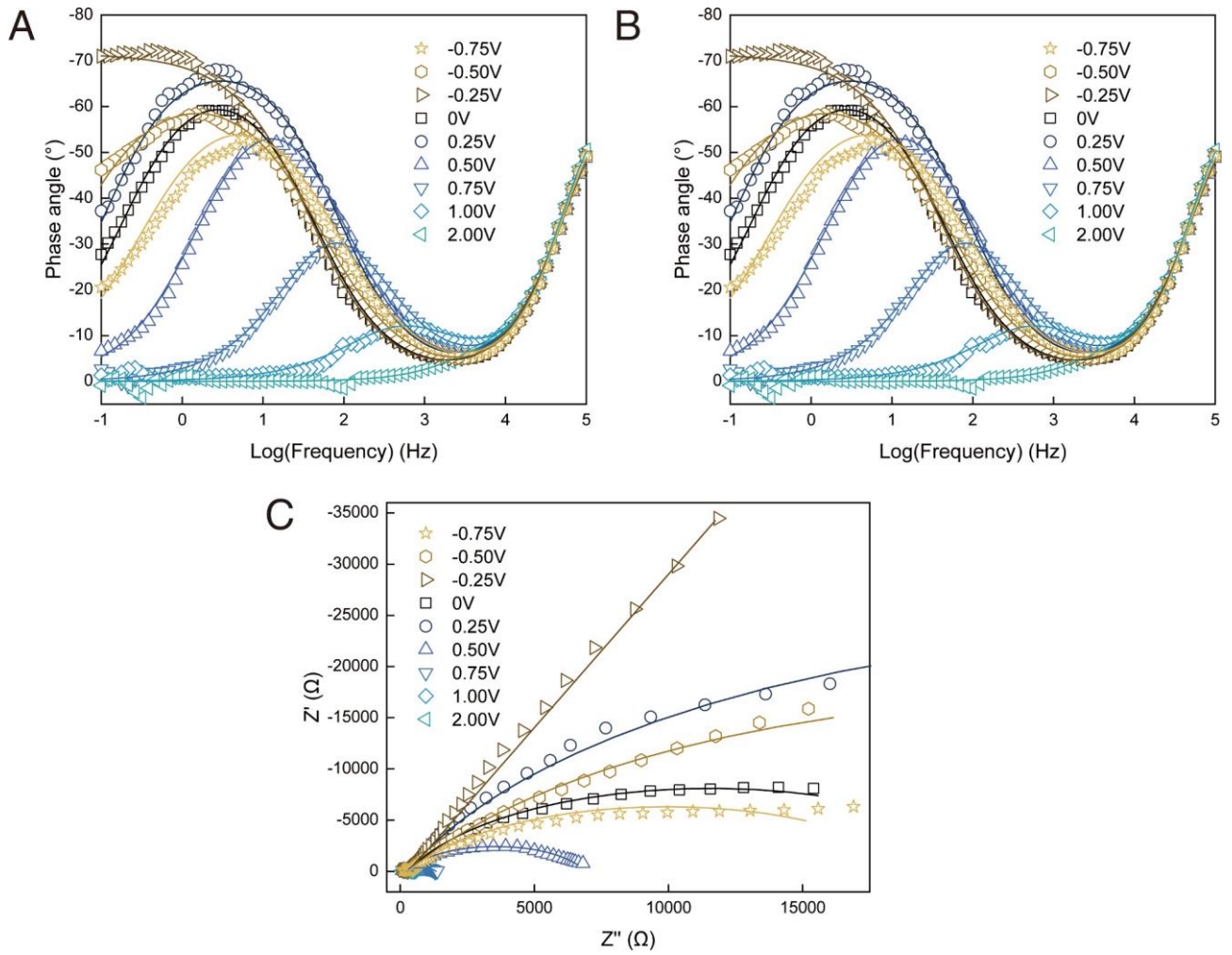


Fig. 5. EIS results at different bias voltages based on open circuit voltage. (A) Bode impedance plots. (B) Bode phase plots. (C) Nyquist plots.

Table 1

Values of electrical parameters obtained by fitting the EIS data for different coating systems during the immersion in physiological saline.

	R_i (Ω)	Q_{EDL}
-0.75 V	1.9E4	3.13E-5
-0.50 V	5.2E4	4.85E-5
-0.25 V	9.0E12	4.01E-5
0 V	6.2E5	4.13E-5
0.25 V	6.3E4	2.14E-5
0.50 V	6.6E3	2.32E-5
0.75 V	1.1E3	1.47E-5
1.00 V	2.3E2	1.52E-5
2.00 V	3.4E1	2.35E-6

3.3. Electrochemical stability of reversible adhesion

To ensure long-term functionality and reliability of bioelectronics devices in wet physiological environment, electrochemical stability of conducting polymers on substrates is one of the most critical requirements. The electrochemical stability and performance of the adhesive layer between KC and PAAm were evaluated using cyclic voltammetry (CV) tests based on physiological saline (Fig. 7A) [20–22]. The CV curves exhibited minimal change across 1,000 charging and discharging cycles, with a decrease in charge storage capability (CSC) of less than 7% after 1,000 cycles, indicating the adhesion's superior electrochemical stability (Fig. 7B).

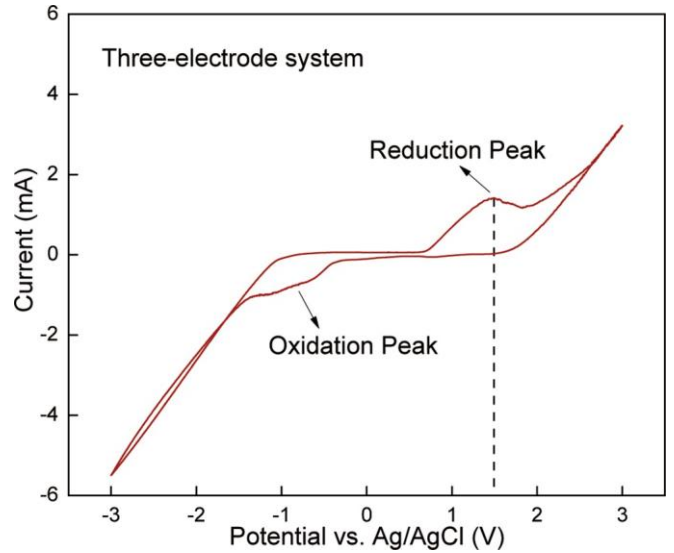


Fig. 6. Electrochemical CV characteristics of adhesive interface.

3.4. Adhesion performance at various voltages

The above CV results demonstrated the stability of electron transfer within the reversible adhesion mechanism. Subsequently, we conducted

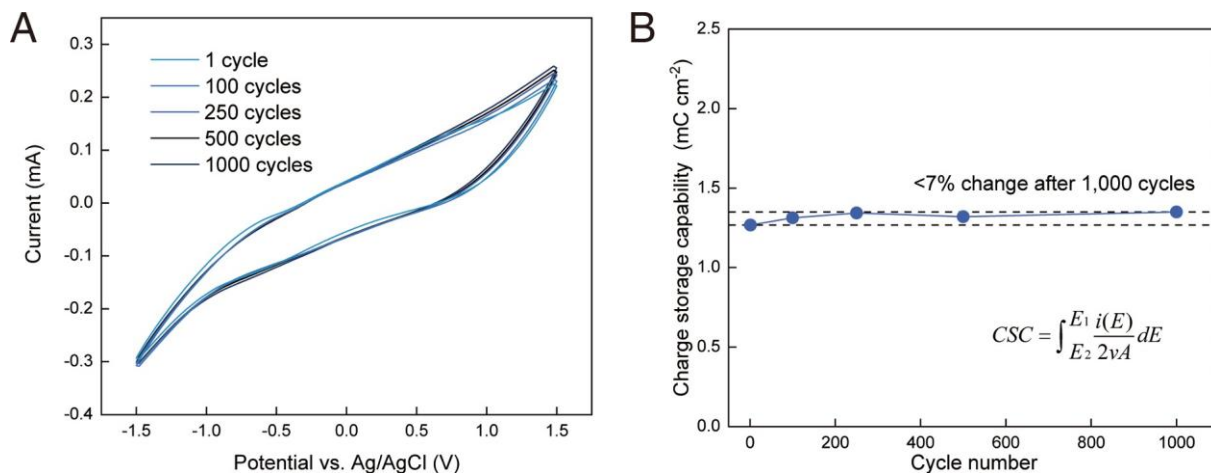


Fig. 7. (A) Long-term CV curves for the adhesive interface between KC and PAAm in physiological saline. (B) Measured CSC versus CV cycle number in physiological saline.

practical assessments to evaluate the reversible stability of the fully swollen hydrogel (Fig. 8). Under positive bias voltage, the adhesion strength can be maintained stably around 7 kPa, while under reverse bias voltage, the adhesion strength is almost non-existent. It is worth noting that under fully swollen conditions, there was a slight increase in adhesion strength (Fig. S4). This is attributed to the increased mobility of ions due to the presence of more water content.

Next, the adhesion strength of the system was evaluated under diverse voltage conditions, exceeding the OER voltage threshold. As Fig. 9 shows, with the voltage increases, the adhesion strength gradually intensifies. However, at voltages surpassing 10 V, failure was observed due to KC fracture, hence adhesion strengths beyond this threshold were not considered. As the bias voltage increases, there is a marked decrease in adhesion strength. Specifically, beyond 6 V, it could be considered that adhesion force is entirely compromised. These voltage-dependent results align with the electrochemical EIS analysis, where increased voltage leads to a reduction in impedance at low frequencies, altering the capacitance value of the ion double layer. Subsequently, we investigated the correlation between adhesion strength and the duration time following electrification. As illustrated in Fig. 10, the adhesion strength exhibited negligible variation over a test duration of up to 5000 s.

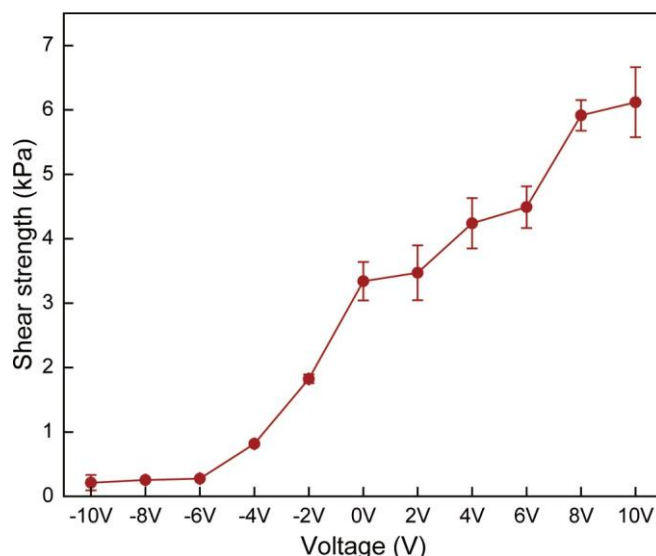


Fig. 9. Shear adhesion strength at different voltages.

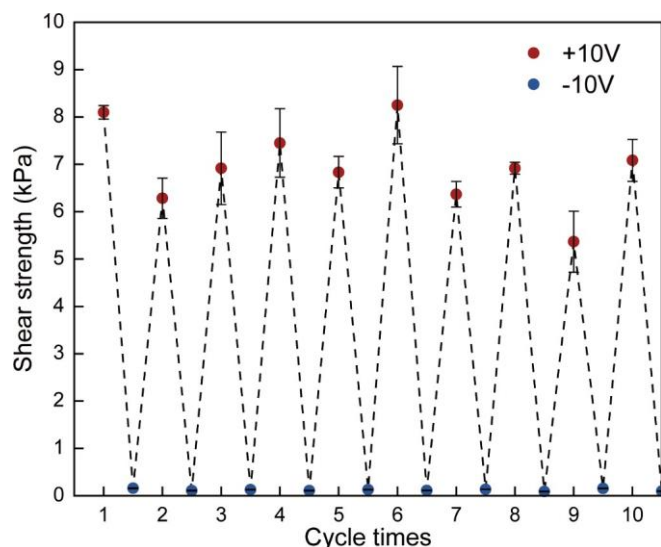


Fig. 8. Cyclic reversible adhesion shear strength test after swelling.

3.5. Reversible electroadhesion applications

We demonstrated reversible electroadhesion pairs that could withstand a shear load of 3.9 kPa at ± 10 V and 0 V, as shown in Fig. 11 and Movie S2. KC was affixed to a carbon electrode, and PAAm serving as the opposite electrode, was connected to a 100 g weight. The contact area between KC and PAAm was 16 cm². Prior to lifting the weight, ± 10 V voltages were applied to the contact interface, followed by the use of a tensile testing machine to lift the weight. When applying a positive bias 10 V voltage, the 100 g weight was easily lifted, whereas in the cases of no voltage and a negative bias 10 V, detachment occurred quickly, failing to lift the weight (Fig. 11 and Movie S2). This demonstrates the effective adhesive strength generated by the described adhesion mechanism.

Next, we demonstrated the biomedical applications of electroadhesion. We applied both electrification and direct adhesion between KC hydrogel and various porcine tissues. We attempted to separate them and subjectively evaluated the adhesion strength, rating it on a scale of 0–4, where 0 = negligible, 1 = weak, 2 = moderate, 3 = strong, and 4 = very strong adhesion. The adhesion results are presented in Table 2. Under direct adhesion conditions, KC exhibited negligible adhesion due to its smooth and low-friction surface. When employing electrified

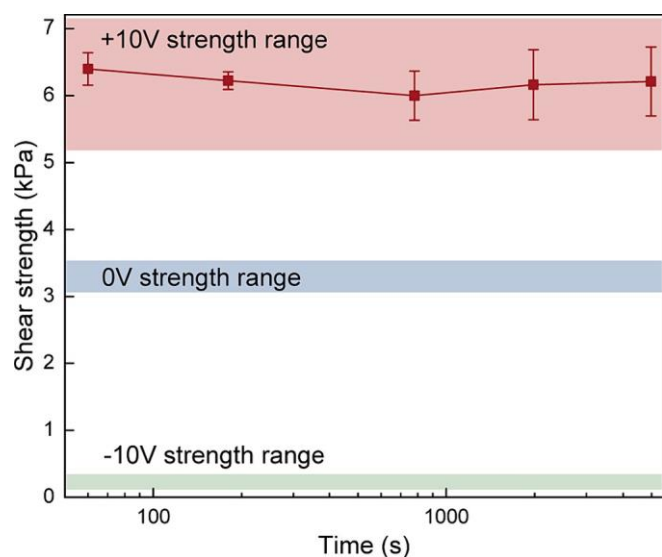


Fig. 10. Curve of adhesion strength with time after electrification. The light-colored range represents the adhesion strength range at 0 s after electrification.

adhesion, a significant increase in adhesion strength was observed for most tissues (except fat). Especially in the case of the heart, aorta, and lungs, detachment from these tissues was particularly challenging. This demonstrates the capability of KC hydrogel to manifest adhesion on certain tissues when subjected to electrified conditions.

4. Conclusions

In this study, we present a way to achieve reversible electroadhesion of hydrogels through low concentration ion migration. We choose κ -carrageenan, which has anionic groups on its molecular chains, and polyacrylamide, which lacks ionic groups and with the solution of physiological saline for reversible adhesion. The reported reversible adhesion has been investigated through electrochemical methods revealing that adhesion arises from the migration of negatively charged molecular chains in KC and cations in PAAm towards the interface and

form ion double layer under positive voltage bias. Conversely, adhesion failure is attributed to the migration of cations in KC and anions in PAAm to the interface under reverse bias, forming ion pairs or even disrupting the double ion layer through mutual migration. Simultaneously, hydrolysis occurs during the electrification process, inducing alterations in the pH value at the interface. This adhesion strategy has resulted in commendable shear adhesion strength and has demonstrated good reversibility (the charge storage capability less than 7% in 1000 times). Subsequently, the adhesion of KC to biological tissues was demonstrated through this strategy. This study provides not only a simple effective method to achieve adhesion but also easily separate. We expect that this study will find extensive applications in areas such as biological tissue adhesion and climbing robots.

Funding

This work was supported by the National Natural Science Foundation of China (51975458, 51605370, and 52375298), the Fundamental Research Funds of Xi'an Jiaotong University for Students (No. 11913223000071), and the Natural Science Foundation of Shaanxi Province (2020JM-010).

CRediT authorship contribution statement

Shan Lu: Writing – original draft, Visualization, Resources, Project

Table 2

Results of electroadhesion tests done with KC hydrogels and various porcine tissues. (Numerical scores represent strength of adhesion assessed on a scale of 0–4: 0-Negligible; 1-Weak; 2-Moderate; 3-Strong; 4-Very strong.).

Tissue (porcine)	Adhesion strength (following electroadhesion)	Adhesion strength (following contact adhesion)
Heart	3	1
Fat	0	0
Aorta	3	0
Lung	2–3	0
Skeletal muscle	1	0
Cartilage	2	0
Skin	1	0

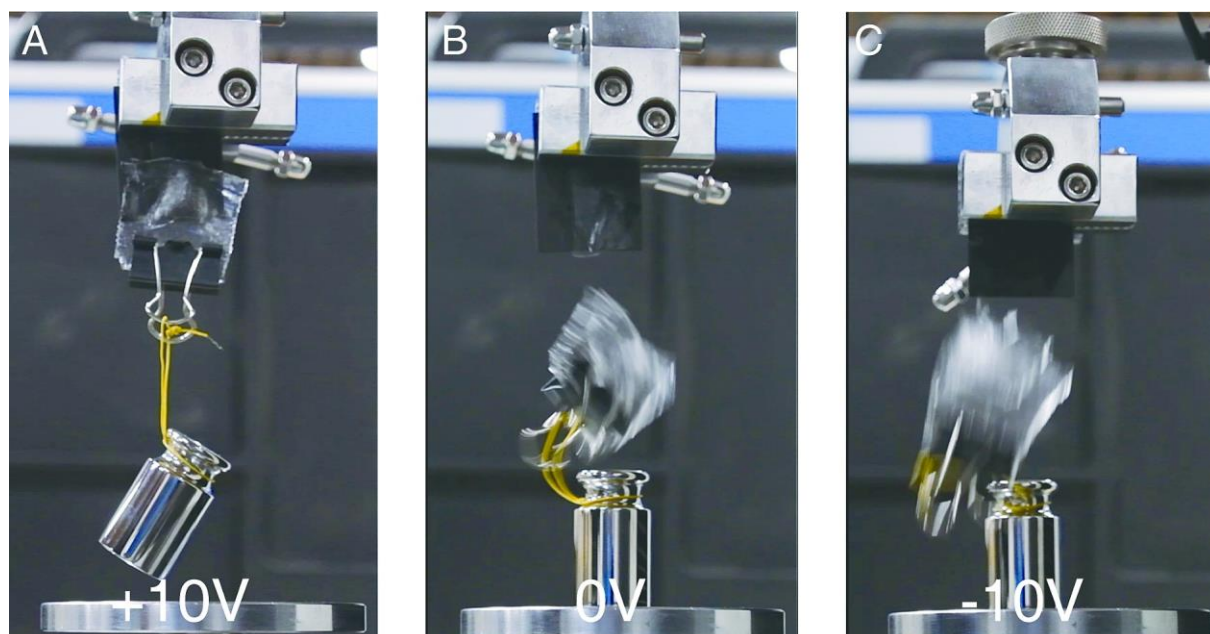


Fig. 11. Demonstration of the use of adhesive lifting weights at different voltages.

administration, Funding acquisition, Data curation, Conceptualization. **Zeyu Ma:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Mingwei Ding:** Data curation, Writing – original draft. **Yuhao Wu:** Writing – review & editing, Software, Resources, Methodology, Conceptualization. **Yuanli Chen:** Methodology, Resources, Writing – review & editing. **Mochen Dong:** Writing – review & editing, Validation, Software, Investigation, Conceptualization. **Liguo Qin:** Writing – review & editing, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ccej.2024.150393>.

References

- [1] H.J. Kim, L. Paquin, C.W. Barney, S. So, B. Chen, Z. Suo, A.J. Crosby, R. C. Hayward, Low-voltage reversible electroadhesion of Ionoelastomer junctions, *Adv. Mater.* 32 (2020) 2000600, <https://doi.org/10.1002/adma.202000600>.
- [2] Y. Gao, K. Wu, Z. Suo, Photodetachable adhesion, *Adv. Mater.* 31 (2019) 1806948, <https://doi.org/10.1002/adma.201806948>.
- [3] M. Liu, L. Jiang, Switchable adhesion on liquid/solid Interfaces, *Adv. Funct. Mater.* 20 (2010) 3753–3764, <https://doi.org/10.1002/adfm.201001208>.
- [4] J. Li, A.D. Celiz, J. Yang, Q. Yang, I. Wamala, W. Whyte, B.R. Seo, N.V. Vasilyev, J. Vlassak, Z. Suo, D.J. Mooney, Tough adhesives for diverse wet surfaces, *Science* 357 (2017) 378–381, <https://doi.org/10.1126/science.aah6362>.
- [5] J.S. Lee, Y.S. Choi, J.S. Lee, E.J. Jeon, S. An, M.S. Lee, H.S. Yang, S.-W. Cho, Mechanically-reinforced and highly adhesive decellularized tissue-derived hydrogel for efficient tissue repair, *Chem. Eng. J.* 427 (2022) 130926, <https://doi.org/10.1016/j.ccej.2021.130926>.
- [6] J. Guo, J. Leng, J. Rossiter, Electroadhesion Technologies for Robotics: a comprehensive review, *IEEE Trans. Rob.* 36 (2020) 313–327, <https://doi.org/10.1109/TRO.2019.2956869>.
- [7] A. Marette, A. Poulin, N. Besse, S. Rosset, D. Briand, H. Shea, Flexible zinc-tin oxide thin film transistors operating at 1 kV for integrated switching of dielectric Elastomer actuators arrays, *Adv. Mater.* 29 (2017) 1700880, <https://doi.org/10.1002/adma.201700880>.
- [8] T.-A. Asoh, W. Kawai, A. Kikuchi, Electrophoretic adhesion of biodegradable hydrogels through the intermediary of oppositely charged polyelectrolytes, *Soft Matter* 8 (2012) 1923–1927, <https://doi.org/10.1039/C1SM06980B>.
- [9] D. Morales, E. Palleau, M.D. Dickey, O.D. Velev, Electro-actuated hydrogel walkers with dual responsive legs, *Soft Matter* 10 (2014) 1337–1348, <https://doi.org/10.1039/C3SM51921J>.
- [10] L.K. Borden, A. Gargava, S.R. Raghavan, Reversible electroadhesion of hydrogels to animal tissues for suture-less repair of cuts or tears, *Nat Commun* 12 (2021) 4419, <https://doi.org/10.1038/s41467-021-24022-x>.
- [11] S. Nam, D. Mooney, Polymeric tissue adhesives, *Chem. Rev.* 121 (2021) 11336–11384, <https://doi.org/10.1021/acs.chemrev.0c00798>.
- [12] S. Lu, Z. Ma, L. Qin, M. Ding, Z. Wang, J. Liu, R. Zhang, Y. Zhang, G. Dong, Effect of molecular helical structure on self-growing and damping of single network κ -carrageenan hydrogel, *Int. J. Biol. Macromol.* 249 (2023) 126082, <https://doi.org/10.1016/j.ijbiomac.2023.126082>.
- [13] M. Diener, J. Adamcik, A. Sa' nchez-Ferrer, F. Jaedig, L. Schefer, R. Mezzenga, Primary, Secondary, Tertiary and Quaternary structure levels in Linear Polysaccharides: from random coil, to single Helix to Supramolecular assembly, *Biomacromolecules* 20 (2019) 1731–1739, <https://doi.org/10.1021/acs.biomac.9b00087>.
- [14] J. Kim, G. Zhang, M. Shi, Z. Suo, Fracture, fatigue, and friction of polymers in which entanglements greatly outnumber cross-links, *Science* 374 (2021) 212–216, <https://doi.org/10.1126/science.abg6320>.
- [15] H. Yuk, C.E. Varela, C.S. Nabzdyk, X. Mao, R.F. Padera, E.T. Roche, X. Zhao, Dry double-sided tape for adhesion of wet tissues and devices, *Nature* 575 (2019) 169–174, <https://doi.org/10.1038/s41586-019-1710-5>.
- [16] Y. Miao, M. Xu, L. Zhang, Electrochemistry-induced improvements of mechanical strength, self-healing, and Interfacial adhesion of hydrogels, *Adv. Mater.* (2021).
- [17] A.R. Narkar, B.P. Lee, Incorporation of anionic Monomer to tune the reversible catechol-boronate complex for pH-responsive, reversible adhesion, *Langmuir* 34 (2018) 9410–9417, <https://doi.org/10.1021/acs.langmuir.8b00373>.
- [18] A.R. Narkar, B. Barker, M. Clisch, J. Jiang, B.P. Lee, pH responsive and oxidation resistant wet adhesive based on reversible catechol-boronate complexation, *Chem. Mater.* 28 (2016) 5432–5439, <https://doi.org/10.1021/acs.chemmater.6b01851>.
- [19] J. Huang, Y. Liu, Y. Yang, Z. Zhou, J. Mao, T. Wu, J. Liu, Q. Cai, C. Peng, Y. Xu, B. Zeng, W. Luo, G. Chen, C. Yuan, L. Dai, Electrically programmable adhesive hydrogels for climbing robots, *science, Robotics* 6 (2021) eabe1858, <https://doi.org/10.1126/scirobotics.abe1858>.
- [20] A. Inoue, H. Yuk, B. Lu, X. Zhao, Strong adhesion of wet conducting polymers on diverse substrates, *Sci. Adv.* 6 (2020) eaay5394, <https://doi.org/10.1126/sciadv.aay5394>.
- [21] M. Ganji, L. Hossain, A. Tanaka, M. Thunemann, E. Halgren, V. Gilja, A. Devor, S. A. Dayeh, Monolithic and scalable au nanorod substrates improve PEDOT–Metal adhesion and stability in neural electrodes, *Adv. Healthc. Mater.* 7 (2018) 1800923, <https://doi.org/10.1002/adhm.201800923>.
- [22] L. Ouyang, B. Wei, C. Kuo, S. Pathak, B. Farrell, D.C. Martin, Enhanced PEDOT adhesion on solid substrates with electrografted P(EDOT-NH₂), *Sci. Adv.* 3 (2017) e1600448.