Strong, Ultrastretchable Hydrogel-Based Multilayered Soft Actuator Composites Enhancing Biologically Inspired Pumping Systems

Hritwick Banerjee, Manivannan Sivaperuman Kalairaj, Hongliang Ren, and Ardian Jusu

Diverse solutions for active fluid movement are known in nature and in human-made devices. However, commercial peristaltic pumps are mostly rigid, non-compliant, and tough to integrate into biocompatible materials. This work aims to approximate actuator-like behavior concerning nonhemolytic pumping action and higher energy density to develop biorobotic physical models and biomedical assistive devices with life-like motion profiles. To achieve this, dielectric elastomers (DEs) offer themselves. DE connected via very high bonding (VHB) tape’s pumping performance is tested and compared to a novel configuration. Comparative analysis of the VHB-based DE pump vis-a-vis the novel design solution involving composite layering of hydrogel and electroactive polymer (HEAP) with interfacial toughness of \( \approx 1522 \pm 188 \) J m\(^{-2} \) exhibits increases in pressure change of up to 68 mmHg at measured flow rates of 16.8 mL s\(^{-1} \) with low viscoelastic losses \(( \approx 8.6 \pm 1.5\% )\) at biaxial prestretch of 3 \( \times \) 3, 10\% stretch rate, and 20 cycles postoperation. The HEAP-sandwiched layer embracing hydrogel-based ionotronics presents 2,205\% ultimate strain and sustains compressive stress of 632 kPa. This pilot thus demonstrates the advantages of greater incorporation of hydrogel-based biocompatible polymers in conjunction with soft active materials and proposes performance characterization for cardiovascular trials and related biofluid pumping applications.

1. Introduction

Nature’s pumping systems allow for a more even stress distribution throughout the circuit than human-made devices.\(^{[1,2]}\) Squid and jellyfish locomotion involves active fluid movement by soft tubular membranes. This also pertains to the transport of fluids within vessel invertebrates and the associated biofluid dynamics. To limit the damage to tissues (e.g., cardiac tissues), it is imperative to bound the problem of local stress maxima. Assistive devices with more muscle-like behavior are now required and soft actuators lend themselves for this purpose.\(^{[3,4]}\) Human-made examples of valveless chamber pumps include bellows and diaphragms, with natural analogies of these found in urethral pumps, jellyfish jet propulsion, dragonfly nymphs’ anal jets, snakes, spiders with venom injection, and insects blood-and-nectar sucking.\(^{[5]}\) Attempts have been made to model the squid and jellyfish with dielectric elastomers (DEs), with limited performance.\(^{[6]}\) Translatory or peristaltic pumping corresponding to valveless moving chambers is found in intestines, mammalian esophagus, anelid hearts, and those of holothurians and arthropods, as well as burrowing worms.\(^{[2]}\) In this spirit, biorobotics aims to emulate natural system performance to study the underlying fundamental mechanisms of complex behaviors such as locomotion\(^{[6,7]}\) in fluids which can involve pumping action.\(^{[8]}\) However, the substantial utilization of soft materials poses an essential discrepancy between animals and conventional robots,\(^{[9]}\) due to its high-impact dissipation energy, damping oscillations, and the smoothing out of irregular movements and forces. Soft active materials are therefore now required to develop compliant, intelligent systems to develop more life-like capabilities with less impedance mismatch vis-a-vis natural systems.\(^{[10,11]}\)

When it comes to soft actuators inspired by muscle-like behavior, a variety of solutions have been proposed, each of which with its limitations. For example, fluidic soft actuators\(^{[12]}\) have been used in a multitude of applications,\(^{[13,14]}\) though the speed of operation and efficiency still pose major limitations. Likewise, thermally actuated fiber-reinforced polymer-based artificial actuators generate large actuation forces, though these solutions are
accompanied by challenges in control and comparatively limited efficiency.[15] DE-based actuators (DEAs) developed by the Stanford Research Institute (SRI)[16] have emerged as one of the promising soft material elastomer-based electromechanical transduction polymers due to their weight, impressive area deformation rate, high energy density, and economical perspective.[17] To that end, Duduta et al.[18] demonstrated DEAs made of strain-limiting elastomers and carbon nanotube electrodes that presented the highest energy density to date. These characteristics put DEAs as one of the potential candidates for pumping operation, as well as for the development of a total artificial heart.[19]

DEAs are commonly constructed using silicone elastomers, acrylic elastomers (e.g., very-high bonding tapes: VHBs from 3 M), polyurethane (PU) elastomers, and natural rubber. Dielectric permittivities and electrical breakdown are two crucial aspects in which silicone dielectric materials underperform compared with acrylic-based DEs. Dielectric permittivities for the acrylic DEs range from 2 to 8, whereas silicone elastomers have dielectric permittivities of $\approx 2$–$3$. Even though silicone elastomers demonstrate superior performance compared with other DEs, their full potential has not been attained as the energy density is low and thus require higher electric fields.[20] Silicone DEs’ actuation strain is usually $<15\%$ when not prestretched, whereas it can reach up to $300\%$ with prestretch provided they survive electrically.[21] Based on these considerations, VHBs are widely used as they are commercially available, come as an adhesive film, can sustain high strain, and are convenient for demonstrating a variety of concepts.[22,23] However, when required to boost the higher frequency response ($\approx 1$–$2$ Hz human heart rate), low damping force, and reduction of viscoelastic losses that closely mimic an efficient blood pump,[24] VHB failed to perform satisfactorily for application. Likewise, biaxial prestretching plays a major role in achieving improved performance with the VHBs 4910/4905 (higher actuation strain and energy density),[25] though there are reports for developing new classes of electropolymer that demonstrate elevated strain without requiring high prestrain.[26]

Interpenetrating networks (IPNs) of VHB 4905/4910 and plasticizing additives are first demonstrated to address instability at interfaces and stress relaxation issues by helping retain prestretch in standalone membranes.[27] In this spirit, Ha et al.[28] also demonstrated that IPN VHB DEs have improved viscoelastic responses and hence elevated operating frequency. Despite significant recent improvements in the performance of electroactive polymers (EAPs), the transfer of these promising technologies for safer human–machine interactions, designing living machines, applications for efficient biohybrid actuation, and sensing remains elusive. In contrast, a mainstay of living machines, applications for electrically driven interfaces, we propose designing tough hydrogel matrices[34,35] with stretchable polymer networks and higher mechanical energy-dissipating network under deformation. In principle, the gel’s toughness is greatly enhanced by introducing energy dissipation in elastic material. Various implementation strategies are widely studied with the given relatively high-level design principle that can serve either as a stretchable network or as a dissipating network.[34,36] A similar synthesis approach is followed to fabricate biocompatible, tough polyacrylamide (PAAm)–alginate hydrogels, as explained in the Experimental Section and in Supporting Information (Table S1 and Figure S1, Supporting Information). Considering escalated hygroscopic properties with a high degree of solubility, lithium chloride (LiCl) solution is prepared at room temperature ($\approx 25^\circ$C) with low relative humidity ($\approx 12\%$) (Table S2, Supporting Information, and Experimental Section). To alleviate DEs’ oxygen reticence effect and expedite ultraviolet (UV)-assisted grafting properties, we treat DE surfaces with 10 wt% benzophenone in ethanol solution (Figure 1a) via swelling-driven surface absorption (Figure S2, Supporting Information, as shown in the study by Yuk et al.[35]). UV-cured hydrogels with LiCl solution (ionogels)[37] were also surface treated with glucose (2 wt%) and glucose oxidase (0.03 wt%) to initiate the process of oxygen scavenging, DE surface absorbed with benzophenone solution at the same time forms an active covalent crosslinking with the surface-treated ionogels to initiate a robust hydrogel and electroactive polymer (HEAP) interface (Figure 1a).

PAAm–alginate hydrogel-based polymer networks act as an elastic solid, whereas water molecules inside behave as an ionic liquid (Figure S3 and S4, Supporting Information). Mobile-ion populations increase substantially when the concentration of LiCl solution increases, with low resistivity demonstrated. High voltage sources are applied at the ends of the HEAP with copper tape and actuated with different voltage values (Figure S5, Supporting Information). DEs are actuated through electrostatic forces. When a voltage difference is induced between the two electrodes, opposing charges accumulate on each electrode. As a result, the attraction force on the DE film causes it to stretch in the radial direction following Maxwell stress (Figure S6 and S7, Supporting Information). Likewise, mobile ions of opposite polarity form an electrical double layer (EDL) inside the HEAP matrix at the interfacial region (Figure 1b).
As shown in Figure 1b, the interlayer porosity (≈15−20 μm) of the ionogels is captured with the SEM. Figure 1c,d shows the pumping mechanism with the multilayered composite structure. A thin layer of the ionogel membrane (≈0.2−0.3 mm thickness) is sandwiched between two VHB 4905 (0.5 mm thickness), as shown in Figure 1c’s inset. Here, pressure sensor and flow meter are embedded externally in series with the HEAP actuator to regulate continuous pressure and flow behavior in the pumping system. When actuated, the multilayer soft actuator expands and fluid flows inside the chamber. When the actuation is taken off, due to elasticity, the pump contracts and hence pushes the fluid upward (Figure 1d).

2.2. HEAP Hybrids’ Mechanical Behavior

To understand the plasticity behavior, mechanical robustness, and ductility of the freshly prepared, transparent ionogels, we conduct uniaxial compression/extension tests utilizing the Instron materials testing machine. Results as shown in Figure 2a suggest that ionogels (sample dimension: 2 × 2 × 2 cm³) have a
uniaxial compression stress of \( \approx 632 \) kPa with an elevated strain of \( \approx 97\% \). Releasing load does not exhibit visible deformation, hence, suggesting high robustness. In situ uniaxial extension test is seen to achieve a maximum strain of \( 220 \)\% with an elevated strain of \( 97\% \). Following the role of pre-stretch in the overall actuator performance,\(^{25,39}\) we measure the critical breakdown voltage of the composite HEAP materials. For biaxial prestretch of \( 3 \times 3 \) VHB 4910, critical voltage is recorded to be \( \approx 6.4 \) kV, whereas HEAP reaches \( \approx 14.7 \) kV (step voltage actuation and increase steps of 50 V each time). This constitutes an increased breakdown voltage of up to \( 296.6\% \). The composite structure, that is HEAP, demonstrates stable oscillation without any viscoelastic losses in between the steps. While the breakdown voltage significantly increased for HEAP, this does not guarantee the same level of performance improvement. It is understood that the thicker the membrane, the higher the breakdown voltage. HEAP possesses a 30\% higher improvement. It is understood that the thicker the membrane, the higher the breakdown voltage.

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mechanical testing. Viscoelastic loss comparison, as shown in Figure 2d, is quantified with the area under the stress–strain curve following normalization. Results shown in Figure 2d further confirm the unprecedented performance improvement with the HEAP (viscoelastic loss [%] at biaxial prestretch of 3 × 3, 10% stretch rate, and 20 cycles postoperation VHB4910: 15.7 ± 1.88; hydrogel: 3.8 ± 0.7; and HEAP: 8.6 ± 1.5 with repetitive trials n = 3–5). Unlike hydrogels with high dehydronation kinetics, mammalian skin epidermis possesses a higher degree of water retention capability that prevents necessary surface biofluid evaporation. Inspired by this skin-like behavior, we present the HEAP with minimal dehydration (1% volume change over 120 h) vis-a-vis ionogel with higher volume reduction (from 100% to 35% of initial volume transition in 120 h), as shown in Figure 2e,f.

Robustness and long-term mechanical stability are prerequisites for any tough, ultra-stretchable gels. In this spirit, we conduct uniaxial tensile tests with 1-week-old ionogels to compare mechanical performance with the freshly prepared ones. 1-week-old ionogel demonstrates an ultimate strain of 219.5% vis-a-vis 220.5% for the fresh samples, further illustrating its mechanical stability (Figure S8, Supporting Information). To recover and extend shelf life, deionized (DI) water solution at 25°C and 6 h of resting time allows the fast swelling ionogel to recover its full structural integrity (100% of the initial volume, Figure S8, Supporting Information). In contrast, HEAP dehydration of 1% over 5 days (120 h) can be recovered in only 5 min of DI water ingestion for functional and morphological symmetry.

2.3. Planar Membrane Performance Comparison

We conduct several planar membrane experiments, as shown in Figure 3a–f, to capture the electromechanical performance of VHB 4910 and HEAP, where both have a comparable thickness of ≈1.3 mm and identical volume. Analytical modeling and finite-element strain theory for the planar membrane are shown in Figure S9 and Figure S10, Supporting Information. Figure 3a–b shows that the breakdown voltage for VHB4910 to HEAP increased by 125% (from 8 to 18 kV). The amplitude of oscillation shown in Figure 3a–b is computed and plotted as a mean of over five cycles of repetition. At 8 kV and a frequency of 0.1 Hz, the VHB 4910 yields a maximum amplitude change of 22.4314 ± 1.3, whereas HEAP at 18 kV and 0.1 Hz shows maximum amplitude deviation (%) of 18.5 ± 1.3. Figure 3c–d, on the other hand, shows an areal strain plot and drift characteristics over 20 cycles. As the applied voltage in Figure 3a–b ramps up from 0 to its peak and then goes down to 0 again to complete one cycle; the areal strain of the actuator increases first to reach its maximum value and then decreases gradually. As the voltage ramps down and returns to zero, the elastomer retains a residual strain due to viscoelasticity. The hysteresis loss after 20 cycles comes as 23.5% for VHB4910 with respect to 18.8% for HEAP. The energy loss (%) measured after the 20th cycle of three biaxial prestretched VHB 4910 at 8 kV is ≈8.851 ± 6.57, whereas for HEAP at 18 kV, it came as ≈40.62 ± 6.5. These performance characteristics provide quantified results of performance superiority of using HEAP over DEs for higher voltage operation.

2.4. Robust HEAP Hybrid Pump Performances

With the possible performance calibration, we conduct HEAP-based wrap-up pumping systems (300% biaxial prestretch) with 200 V sec⁻¹ ramp rate and 20 cycles of operation with a modulated square wave. Figure 4a–b shows an average flow rate behavior of 16.8 mL s⁻¹, whereas pressure change of up to 68 mmHg is observed around 0.25 Hz frequency of operation. During frequency sweep from 0.1 to 1.5 Hz, the HEAP-based pumping system exhibits its system resonance of ≈0.25 Hz of pumping action. During this frequency regime, maximum electric energy transfers into hydraulics and hence maximum natural pumping system oscillation. As the electric field surpasses a certain threshold, mobile electrons then can tunnel from the cathode surface into the ionogel-sandwiched medium. Thereafter, the pumping pressure P increases with the square of the electric field E and linearly with the dielectric constant (ε) of the composite materials as $P = k_\text{fl} E^2$, where $k_\text{fl}$ demonstrates a constant depending on the morphology/dimension of the pump. Generated flow rate $Q$ has a relation with the electric field squared and is proportional with the fourth power of the effective channel size, $H$: $Q = k_\text{Q} E^2 H^4$. $k_\text{Q}$ presents a proportionality constant based on the pump dimension. Nominal electric field intensity of 17 MV m⁻¹ is quantified before electroactuation and morphological deformation. With biaxial prestretch of 2 × 3 applied to the HEAP membrane, the electric field intensity observed is in the range of 95–98 MV m⁻¹. The electric field intensity decreases slightly due to the additional stretch imposed by the fluid pressure within the membrane. This value of the electric field intensity is measured before any deformation by the membrane with spatially and temporally varying deformation during pumping.

Based on a DE-based pump comparison, to the best of our knowledge, HEAP is found to achieve the highest pressure change measured to date. Based on the results, as shown in Figure 4a,b, we find the system proposed to achieve 56.7% pressure range of the systolic human heart condition. The oscillation profile appears to be consistent, as obtained with multiple trials, (n = 5) and mean, as shown in Figure 4c. The HEAP-based peristaltic pump can conduct fairly cyclic behavior over multiple repetition cycles. A slight drift of amplitude as ≈4.7% over nine cycles of oscillation is shown in Figure 4c.

To quantify the robustness of the HEAP-hybrid pump performance, we demonstrate 1000 cycles of pumping performance, as shown here in Figure 4d. Peak flow rates are measured to be 17 ± 0.2 mL s⁻¹, 16.2 ± 0.4 mL s⁻¹, 15.9 ± 0.5 mL s⁻¹, 15.7 ± 0.7 mL s⁻¹, and 15.4 ± 0.95 mL s⁻¹ for the 1st, 10th, 100th, 500th, and the 1000th cycle, respectively. Experiments are conducted at least three times and data presented as mean ± standard deviation with fresh HEAP samples. The initial triggered flowrate changes comply with the HEAP membrane’s transient viscoelasticity nature that transforms into a steady state with more oscillation cycles. Here, 9.41% peak flow rate change is observed for 1000 cycles of pumping action, further demonstrating potential applications.

We aim to address the next step toward a more robust HEAP pumping for an increased number of cycles of operation to justify its niche biomedical applications. With the related literature
Figure 3. Comparative analysis and performance monitoring of the DE and HEAP with different parameters. a,b) Amplitude of oscillation for VHB 4910 and HEAP with different actuation voltages and frequencies of operation, respectively. c,d) Areal strain with multiple cycles of oscillation (20 cycles) for VHB 4910 and HEAP, respectively, before snap through. e,f) Energy loss for VHB 4910 and HEAP with different actuation voltages and frequencies of operation, respectively.
taken into consideration, the flow rate that has been achieved with the HEAP multilayer composite materials is the third highest in the DE-based composite pumping system to date, as shown in Figure 5. It is anticipated that the flow rate can be further improved upon with an arrangement of two HEAPs working in tandem.

3. Application Arena and Rationale

Potential applications of the HEAP-based biologically informed pumping systems include cardiovascular and renal assistive devices, and testing of compatibility via animal models (Figure 6). Moreover, in the near term, we propose to design sandwiched layers for multistimuli-responsive soft sensors and integrate them with HEAP-based pumping systems for diverse intelligent, biohybrid robotic applications. The physiologically relevant applications of hydrogel-encapsulated soft active materials also include soft exosuits, as well as veterinary biometric monitoring, diagnostics, and support. Functional nanomaterial composites for soft sensing can potentially help with biomechanics research, diagnostics in veterinary care, and animal welfare more broadly.

3.1. Cardiac Assist Devices (CADs)

A healthy human heart beats 72 times a minute at a stroke volume of 70 mL per beat that results in 5 L min⁻¹ average adult cardiac output. Moreover, taking into account human pulse rate between 40 and 120 beats per minute, the equivalent frequency comes around 0.6–2 Hz. HEAP-based pumping systems, as demonstrated in this article, can achieve a flow rate of 1.08 L min⁻¹ whereas the maximum oscillation frequency was set as 1 Hz. A
similar pressure trend is observed as healthy human heart systolic/diastolic pressure is around 110/70 mmHg. HEAP-based multimaterial pumping system operates optimally around 68 mmHg. These results present an exciting near-term opportunity to step up for all soft, biocompatible heart valve replacements.

To assist failing hearts, commercial blood pumps are frequently used that pose a threat to patients’ overall well-being and recuperation. Existing problems with the centrifugal blood pumps are friction between shaft and seal that causes heat generation, which strongly correlates with hemolysis. In roller pumps, at points where rollers narrow, the pipe leads to a thin boundary layer with a high-gradient friction causing high shear stresses. The rate for blood damage for roller pumps is estimated to be $4 \times 10^{-4}$ mg $\text{dl}^{-1}$ $\text{s}^{-1}$. More information related to commercially available CADs and their properties is shown in Table S3, Supporting Information. A blood pump using novel soft active materials is thus an obvious choice. Future applications of such a fluid pump include an implant as a ventricular assist device (VAD). Similarly, the technology can be extrapolated to other biomedical arenas, such as treatment for intrauterine growth retardation (IUGR), a placental vascular disease that results in insufficient nutrient transfer to support normal fetal growth. Specifically, a novel wrap pump made up of soft active materials may be used to improve flow through a series of contractile motions applied on the umbilical cord, as shown in Figure 6. It may similarly be applied to an artery to improve blood flow.

### 3.2. Postrenal Failure Assistance

The human body can have hypervolemia or hypovolemia due to fluid imbalance. During robot-assisted partial nephrectomy and robotic transabdominal kidney transplantation, there is an unmet need to mechanically pump urine out to maintain the body fluid level in a minimally invasive way. As a proof of concept, we build a miniature version of a HEAP-based pump that is mimicked as wrapped in the ureter to pass the residue fluid by changing the actuation force, as shown in Figure 7. A soft, flexible tube (TYGON ultra soft tubing) of 250 mm long, with outer diameter of 4 mm, and inner diameter of 3 mm is used and the HEAP membrane is placed midway along the length of the tube. The HEAP membranes shown in Figure 7a,c are equipped with five concentric layers to increase the maximum wall pressure as opposed to a single layer. The final dimension of each concentric HEAP membrane is 4 mm long, the outer diameter of 10.5 mm, and inner diameter is 4 mm to fit closely with the simulated ureter. Figure 7b shows the HEAP pumping system’s actuation mechanism, where each electroactuation spanned for 2 s. We use a low actuation voltage of 1.5 kV for the biomedical engineering safety constraint, hence less deformation. When miniaturized HEAP membrane is relaxed, it compresses the flexible tube laterally, flushing out the simulated red body fluid; while electroactuated, HEAP membrane expands, resulting in fluid flow (Figure 7c). For one HEAP membrane, the steady-state flow rate is measured as 0.4 mL min$^{-1}$, whereas for three HEAP membranes in tandem, 1 mL min$^{-1}$ steady flow rate is observed. Due to HEAP

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**Figure 5.** Soft electroactive polymer actuator-based pump performance comparison: flow rate and the pressure change reported in this work and in the recent related literature.[41,53–58]

**Figure 6.** Potential applications, a) biomedical applications including ventricular assistive devices, IUGR, and assisting urinary device for acute renal failures and artificial urinary sphincter and b) integrative biological applications for comparative physiology and physical modeling of the biomechanics of jellyfish, octopus, squid, and puffer fish-inspired pumping.
membranes’ actuation sequence, the flow rate increase up to 150% transitioning from one HEAP to three HEAP membranes in tandem (Figure 7b and Movie 1, Supporting Information).

We propose to progress further to design a biocompatible soft wrap-up pumping system that will accommodate an implantable energy source, the cyclic procedure of inflation/deflation to support weak kidney patients. HEAP-based pumping can also work as an adaptable, customized stiffness manipulator for clutching ureter tissues, manipulating cellular aggregate in the human body, and optimal drug delivery for enhancing patients’ life.

3.3. Neuromechanics and Integrative Biology

The ultimate goal of intelligent materials is to achieve adaptive behavior in rapidly changing, irregular environments, as exemplified by nature. Bionics and biomimicry-related research has enjoyed steady growth in the past two decades.\(^{49,50}\) Greater insight into the underlying mechanisms of animals’ adaptive behavior is a prerequisite for reverse engineering them by abstraction of the salient features, rather than by an undirected attempt at mimicry. This not only pertains to unfocused mimicry but also the realization of increasingly life-like robot behavior under the implementation of soft actuators and sensors.

Experimental robotics, in turn, allow for greater parameter sweep due to more control over inputs than in biological experiments, thus providing an opportunity to generate new hypotheses. Outputs can also be measured at high resolution and fidelity. To further push the frontier of neuromechanics and integrative biology, there is an unmet demand to transport fluids within invertebrate vessels and the associated biofluid dynamics. Direct motivation in building and assisting soft active pumping systems comes from the jellyfish jet propulsion, dragonfly nymphs’ anal jets, spiders with venom injection, insects blood-and-nectar sucking, ommastrephidae, and many others. We foresee using a HEAP-based pumping system to emulate marine biology to mimic these incredibly captivating biophysical aerodynamics with bipedal phase transition. We believe that HEAP-based pumping systems can harness gaps in propulsion by introducing a much higher pressure change and flow rate behavior.

4. Conclusion

In summary, in this article, we present novel groundwork in the application of HEAP as a fluid pump, introducing advanced soft composite materials manufacturing techniques. HEAP
5. Experimental Section

Materials: LiCl (VWR Chemicals; ≥ 99.9%) was used for its high hydroscopic capability and for elevating PAAm—alginate pregel solution’s ionic conductivity. Dichloromethane (DCM) (Sigma-Aldrich; ≥ 99.9%), acetone (Sigma-Aldrich; ≥ 99.5%), ethanol (Sigma-Aldrich; ≥ 99.5%) and, methanol (Sigma-Aldrich; ≥ 99.9%) were used as received without additional modification. Acrylamide (AAm) monomer and LiCl powder were dissolved in deionized water. Glucose (Sigma-Aldrich; ≥ 99.5%) and glucose oxidase (Sigma-Aldrich; ≥ 99.5%) were used as an oxygen scavenger in hydrogels for the effect of elastomer surface treatments. VHB DE membranes were fabricated using VHB 4910 and VHB 4905 (3M Inc.), whereas in all DE membrane types, carbon grease (MG-846-80G; MG Chemicals) was applied on both sides of the DE membrane as soft electrodes using a simple paintbrush. Benzophenone (Sigma-Aldrich; ≥ 99%) was introduced for DE surface treatment to form an oxygen-inhibited layer. Components from National Instruments were used for signal processing within the measurement setup, which was controlled by LabView 2012.

PAAm—Alginate Hydrogels Synthesis: The PAAm—alginate hydrogels synthesis was initiated with ionically crosslinking sodium alginate and covalently crosslinking acrylamide with customized ingredients and concentration in one-pot stock solutions. One-pot stock solutions were then passed onto ultrasonication (Sonica 5200) for 4 h with continuous stirring and left overnight for crosslinking. Chemicals used to produce PAAm—alginate hydrogels were acrylamide (AAm, Sigma-Aldrich; ≥ 99.9%), sodium alginate (SA, Sigma-Aldrich; ≤ 15.5%), 2-hydroxy-4’-(2-hydro-2-methylpropionyloxy)benzophenone (photoinitiator or PI, Sigma-Aldrich; ≥ 97.5%), and N,N’-methylenebis(acrylamide) (MBAA, Sigma-Aldrich; ≈99%). The percentage of the weight used for fabricating strong, ultra-stretchable PAAm—alginate hydrogel was AAm of 22.6%wt, SA of 1.5%wt, PI of 0.58%wt, MBAA of 0.020%wt, and DI water (Milli-Q pore; 18.2 MΩ) of 75.3%wt UV treatment for 2 h followed by a day resting time.

Hydorgels’ Tough Bonding onto DEs: To form a stable hydrogel-based ionotronics with optimized mechanical robustness, AAm concentration was fixed at 2.2 M, and LiCl’s concentration was calibrated to be 1.3 M. This method to enhance the electromechanical deformation with improved ionic conductivity was based on a previously reported protocol[16,17] where, freshly prepared PAAm—alginate hydrogels were added into LiCl powder for stretchable ionics. DE surfaces were treated by absorbing benzophenone to enhance the effect of oxygen inhibition.[15] Benzophenone solution (10 wt% in ethanol) was evenly applied onto the DE surfaces and washed thoroughly multiple times with methanol and then completely dried using nitroel gas. In freshly prepared ionogel (PAAm hydrogel – LiCl), 2 wt% of glucose and 0.02 wt% of glucose oxidase were added as an oxygen scavenger. Thereafter, the physically cross-linked ionogel solution was transferred into the surface-treated DE membrane. Ionogel—DE membrane hybrid was then UV (XYZ Printing) treated with 60 W, 375—405 nm wavelength for 2 h, and rested for a day in a humidified chamber before use.

SEM: PAAm—alginate hydrogel-based ionogels were fast frozen at −80°C and then dried in a freeze dryer (Virtisfreezemobile, VirtisCo., Gardiner, USA), and the surface morphology and porosity were investigated using SEM (FEI Quanta 600andFESEM; JEOL-JSM-6610LV) operating at 15.0 kV for low- and high-resolution imaging.

Tensile Testing: The 90° peeling test, stress—strain analysis, and the cyclic test for hysteresis and viscoelastic calibration were conducted using an uniaxial testing machine in Instron 5543 (Illinois Tool Works Inc., USA) with 1 kN load cell at a ramp rate of 2 mm min⁻¹.

Designing and 3D Printing: The final design of the wrap-up pump was designed using Solidworks (Dassault Systèmes, SolidWorks Corporation) and 3D printing underwent using Projet MJP 5600 (3DS).

Spin Coating: We used a spin coater (Agent, WS-650-23NPP) for 60 s with 2000 rpm speed for fabricating a thin layer of ionogel membrane as the sandwiched layer.

Multilayer Soft Actuator-Based Pump Manufacturing: Adhesive tape of 0.5 mm was stretched biaxially and then pasted onto the acrylic frame. Next, a short copper tape was pasted from the rectangular region to the outside of the frame. Carbon grease electrodes were then painted first on the top side of the film. After that, another layer of 3M VHB 0.5 mm tape was stretched and placed again on top of the painted electrode, sandwiching the electrode between two tape layers. The frame was then flipped and another electrode was painted onto the 3M VHB tape. Excess material was cut off from the sides of the frame using a penknife. Finally, the wrap pump layer was wrapped around the tubular frame by rolling.

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

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