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Soft iontronic delivery devices based on an intrinsically stretchable ion selective membrane

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Abstract

Implantable electronically controlled drug delivery devices can provide precision therapeutic treatments by highly spatiotemporally controlled delivery. Iontronic delivery devices rely on the movement of ions rather than liquid, and can therefore achieve electronically controlled precision delivery in a compact setting without disturbing the microenvironment within the tissue with fluid flow. For maximum precision, the delivery device needs to be closely integrated into the tissue, which is challenging due to the mechanical mismatch between the soft tissue and the harder devices. Here we address this challenge by developing a soft and stretchable iontronic delivery device. By formulating an ink based on an in-house synthesized hyperbranched polyelectrolyte, water dispersed polyurethane, and a thickening agent, a viscous ink is developed for stencil patterning of soft ion exchange membranes (IEMs). We use this ink for developing soft and stretchable delivery devices, which are characterized both in the relaxed and stretched state. We find that their functionality is preserved up to 100% strain, with small variations in resistance due to the strain. Finally, we develop a skin patch to demonstrate the outstanding conformability of the developed device. The presented technology is attractive for future soft implantable delivery devices, and the stretchable IEMs may also find applications within wearable energy devices.

1. Introduction

Stretchable electronics allow for seamless integration of electronics with the human body. Recent progress has yielded stretchable components like batteries, light emitting diodes, opto-electronic devices, supercapacitors, sensors, and biomedical implants [1, 2]. While a plethora of stretchable devices have been developed for electrical interfacing of tissue [3–5], it would be beneficial with multimodal devices that also allow for simultaneous chemical stimulation. To date, microfluidics have been integrated into stretchable electronic systems for drug delivery [6]. Although versatile [7–9], implanted fluid delivery systems come with many challenges and limitations, including bulky pressure actuating systems, local pressure and fluid control, and limited spatial and temporal resolution. An alternative technology that would be attractive for stretchable multi-modal biomedical devices is iontronic delivery devices [10, 11]. These devices are based on electrophoretic transport

of charged substances through ion exchange membranes (IEMs), often driven by conducting polymer electrodes. Iontronic delivery devices exhibit several attractive features including fluid-free delivery, absence of moving mechanical parts, and high spatiotemporal resolution [12–18]. IEMs are one of the critical components within iontronic delivery devices and several different materials that have shown excellent ion selective properties have been developed, including oxidized polycationic poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) [10], poly(2-acrylamido-2-methyl-1-propanesulfonic acid) [19], poly(styrene sulfonate)-co-maleic acid cross-linked with polyethylene glycol (PSS-co-MA/PEG) [20], and polyanionic poly(diallyldimethylammonium chloride) [19], poly(vinylbenzylchloride) quaternized by dimethylbenzylamine [21], and poly(vinylbenzyl chloride) quaternized by triphenylphosphine (PVBPPH3) [22]. Although some degree of flexibility has been achieved in, e.g. screen printed ion pumps on poly(ethylene

terephthalate) (PET) substrates [23], the high degree of crosslinking within these IEM materials has prevented the development of stretchable devices. By overcoming this limitation, novel applications including drug delivery to lungs, heart, muscles, joints and brain would be possible [22].

Recently, there has been major progress in the development of stretchable and tough hydrogels [24]. The key mechanism behind this progress is the combination of strong and weak crosslinkers by the incorporation of dual polymeric networks, often with sacrificial crosslinking bonds, that strengthen the hydrogels, making them less brittle than conventional single-network hydrogels. However, these hydrogels cannot be directly applied for the envisioned applications as they are not ion selective. Recently, the first stretchable IEM materials were developed for ionic diodes. Lee *et al* [25] UV-co-polymerized two polymer networks to achieve the materials while Wang *et al* [26] blended polyelectrolytes into UV polymerized polyacrylamide gels and manually cut out pieces of the materials. Although promising materials, the above processing methods are not ideal for the fabrication of biomedical devices. The UV polymerization approach by Lee *et al* used pico-injection of precursor solution and subsequent removal of unreacted solution. The cut and place approach employed by Wang *et al* is not very scalable and does not allow for miniaturization. Also, UV polymerization approaches need to consider the possibility of unreacted monomer residues within the materials.

Here we report the first stretchable iontronic delivery device based on soft IEM channels embedded in polydimethylsiloxane (PDMS). A printable ink, based on an anionically charged hyperbranched polyglycerol (A-Hpg) dendrolyte [27], water-dispersed polyurethane (WPU), and 2-hydroxyethyl cellulose (2-HEC), was formulated for the fabrication of the stretchable IEM channels. Our in-house synthesized A-Hpg polyelectrolyte was used due to its good ion selectivity and ability to transport large ions [28, 29]. The film was made stretchable by the incorporation of a WPU network that formed upon drying of the ink. The printability of the ink was achieved through the incorporation of 2-HEC, which resulted in an ink within the rheological requirement for stencil patterning. The stretchable IEMs were characterized with respect to selectivity, resistance, and stretchability, showing good performance in all aspects. Finally, to show the feasibility of these iontronic devices in stretchable applications, we demonstrated a device mounted on the finger while delivering protons during bending-induced stretching.

2. Experimental

2.1. Materials and processing

The ink for the stretchable membrane was based on an in-house synthesized A-Hpg [28], whose

chemical structure consists of hyperbranched polyglycerol ($M_w \sim 10\,000$ Da, degree of branching $\sim 55\%$ – 60% , polydispersity index (PDI) ~ 1.6) covalently functionalized with 80% anionic butylsulfonate groups and 20% polymerizable unsaturated allylic groups. The A-Hpg powder was dissolved in deionized (DI) water to 50 wt.% concentration by vortexing to form a homogeneous A-Hpg solution (figure 1(b)). The A-Hpg solution was then mixed with a water-based polyurethane dispersion (U4101 Alberdingk Boley, 39–41 wt% solid content) in a 55:45 volume ratio. As the obtained mixture had too low viscosity for a stencil patternable ink, 2-HEC (Sigma-Aldrich $M_w \sim 380\,000$) was added to a 5 wt% concentration and mixed with a planetary centrifugal mixer for 10 min to achieve a viscous white mixture with rheological properties for stencil patterning. Figure 2(a) shows a schematic of the fabrication process. A 2 inch silanized (trichloro (1H,1H,2H, 2H-perfluorooctyl) silane) glass wafer was used as a supporting substrate for the device fabrication. PDMS (Sylgard 184, Dow Corning, 10:1 ratio) was spin coated at 16 rps for 30 s on the glass wafer to achieve a film thickness of $80\ \mu\text{m}$ (step 1) and was then cured on a hotplate at $80\ ^\circ\text{C}$ for 3 h (step 2). The PDMS substrate was oxygen plasma treated for 4 min at 70 W to induce a hydrophilic surface [30]. The IEM ink was stencil patterned manually onto the PDMS surface through a laser patterned $125\ \mu\text{m}$ thick polyethylene terephthalate sheet (Polifoil Bias, Policrom Screen) (step 3). The print was cured in an oven at $80\ ^\circ\text{C}$ for 12 h, resulting in a $60\ \mu\text{m}$ thick IEM pattern (step 4). The middle portion of the IEM channel was encapsulated by spin coating of PDMS at 16 rps for 30 s through a stencil mask made of manually cut $25\ \mu\text{m}$ thick poly (ethylene naphthalate) foil (Teonex) (step 5). The top PDMS layer was finally cured at $80\ ^\circ\text{C}$ for 3 h (step 6). The encapsulation layer both prevented ions from escaping the channel and reduced the drying rate of the hydrated channels. Individual devices were manually cut out from the supporting glass wafer for electromechanical characterization. Freestanding stretchable IEM films for mechanical characterization were made by pouring the stencil patternable ink into a rectangular Teflon mold followed by curing in an oven at $80\ ^\circ\text{C}$ for 12 h.

2.2. Device characterization

All devices were immersed in 10 mM potassium chloride (KCl) (aq) for at least 12 h prior to testing. Electrical characterization was performed using a Keithley 2600 series source meter controlled by a custom LabVIEW program. Stretching experiments with electrical characterization were performed using a motorized linear stage (Zaber X-LSQ300A-E01) at $2\ \text{mm s}^{-1}$ stretching speed. Electrical measurements were performed by sourcing a 500 nA current while measuring the voltage. A 10 mM KCl

(aq) electrolyte was used in both source and target reservoirs for all the stretching measurements. Ag/AgCl wires (Ag wires, Sigma-Aldrich, immersed in ferric chloride solution for 5 min) were used as electrodes for electrical measurements. Stress-strain tests were performed on freestanding 0.9 mm thick and 3.6–4 mm wide membrane samples. Samples were kept immersed in 10 mM KCl for 24 h before

stretching to 25%, 50%, 75%, 100%, and maximum strain consecutively ($n = 4$) at 0.2 mm s^{-1} while the force was measured with a force gauge (Mark-10, M5-012, 500 mN range). The Young's modulus was calculated at 10% strain from the slope of the averaged ($n = 4$) stress-strain curve. To avoid channel dehydration during the experiment, all the measurements were performed in well hydrated

environment crafted by DI water immersed clean room tissues. The clean room tissues were placed right below and on the sides of the devices during characterization.

3. Results and discussion

The chemical structure of the constituent materials of the stretchable IEMs are shown in figure 1(a). The IEM component is based on A-Hpg, which is an excellent material for transport of biologically relevant substances that are too large for many other IEM materials (PSS, PSS co-MA, PVBPh₃) [27]. To induce mechanical robustness and stretchability to the membranes, a water-based polyurethane (WPU) dispersion was added to the formulation. A similar concept has previously been used to induce outstanding stretchability in PEDOT:PSS films, as the WPU in that case forms a percolating stretchable network within the composite [31]. Finally, to achieve a stencil printable ink, the thickening agent 2-HEC, a gelling and thickening agent derived from cellulose [32], was added to enhance the printability of the ink. As all the components are dispersible or soluble in water, stretchable membranes can be easily formed by stencil printing or drop casting from an aqueous mixture of the components. Upon drying, the WPU forms a water insoluble network within the composite which provides the mechanical robustness of the IEM (figure 1(c)). The A-Hpg network created in the cured composite is responsible for the ion selectivity in the formulated membrane. Freestanding stretchable IEMs were fabricated by drop casting and subsequently stretched in the wet state to 25%, 50%, 75% and 100% strain (figure 1(d)). The calculated Young's modulus was 41 ± 3 kPa for the freestanding IEM (at 10% strain), which is well below that of the encapsulating PDMS in the iontronic device (MPa range) [33]. The PDMS encapsulation is therefore dominating the mechanical behavior of the stretchable device. The freestanding membranes exhibited a viscoelastic behavior when stretched and recovered close to the original length upon release, e.g. for 100% strain the samples recovered to 125% of their original length (figure 1(e)). The maximum strain of the tested freestanding membranes was in the 200%–350% range (figure 1(e)). The variability in maximum strain of these freestanding films (see figure 1(e)) most likely came from the mold-based fabrication approach for producing mm-thick films. The approach caused the formation of larger clusters of WPU, which was not observed in the stencil printing process used for fabricating the stretchable iontronic devices.

After formulating the stretchable IEM material, the next step was to integrate it into an iontronic delivery device, the electronic ion pump [10–12], which comprises two reservoirs with an encapsulated IEM channel in between. PDMS was chosen as the

substrate material as it is widely used in microfluidics and is transparent, nontoxic, inert and has excellent viscoelastic properties similar to tissues [34, 35]. The ion pumps were fabricated according to the steps illustrated in figure 2. To verify the functionality of the stretchable ion pumps, their transport selectivity was studied. The IEM channels comprised of anionically charged A-Hpg, which should facilitate cation dominated transport through the channel. To evaluate the ion transport, a voltage was applied between the source and the target reservoirs using Ag/AgCl electrodes, with the positive electrode connected to the source and the negative connected to the target. The measured total current is the sum of the cationic and anionic current. For a cation selective membrane, cation transport should dominate, and the current should scale with the diffusion coefficient of the transported cationic species in the target reservoir. To evaluate this correlation, we tested three different source electrolytes with the same anions: acetylcholine chloride (AChCl), sodium chloride (NaCl), and KCl. The conductivity of the channel was measured under constant voltage while changing the source electrolyte composition. The results in figure 3(a) show that the conductivity scales linearly with the cation diffusion coefficients [36], as expected for an IEM channel. To further study the selectivity of the transport, we kept the source electrolyte composition constant (KCl) while switching the target electrolyte between potassium acetate (KAc), potassium perchlorate (KClO₄), and KCl. The measured conductivities show no dependence on the target electrolyte composition (figure 3(b)), indicating good selectivity with little backflow of anions from the target. Finally, the dependency of the channel conductivity on the source electrolyte was measured (figure 3(c)), showing only a weak dependence on the source concentration. To demonstrate the ion transport capability of the device, H⁺ was transported from the source reservoir (10 mM HCl) to a target reservoir comprising the pH indicator methyl red (figure 3(d)). After 15 min of operation, the color of the target reservoir had shifted from yellow to red, indicating effective transport of H⁺. Altogether, these studies confirm that the developed ion pump is functional in the non-stretched state.

To study the operation of the stretchable ion pump under strain, the device was mounted in a linear stretching setup (figure 4(a)). The ion pump was operated at a constant current of 500 nA with KCl electrolytes and the resistance of the channel was calculated from the measured voltage. The device was stretched for five cycles to 25%, 50%, 75%, and 100% strain, with 30 s resting periods at the targeted strain levels and at 0% strain. Figure 4(b) shows the relative resistance for a devices during this stretching sequence. The relative resistance increases with increasing strain, as expected. Figure 4(c) shows the extracted relative changes in resistance versus strain

from each cycle, demonstrating that the device performs well even at 100% strain. One should note that even though the relative changes in figure 4(c) are regular, a long-term drift in both the relaxed and strained state can be observed in figure 4(b) for higher strains. This kind of behavior is not uncommon for electronic conductors, as irreversible changes occur under strain in the percolating network. For ionic conductors, however, such behavior is less common due to the softer and deformable nature of the ionic phase. In this case, we suspected that the resistance drift at higher strains was caused by slight dehydration of the IEM channel. To investigate this, a strain cycling experiment was performed without providing a humid environment. Figure 4(d) shows a substantial increase in resistance with time, especially at elevated strains, reaching more than five times higher resistances at the end of the strain cycles in comparison to the more humid conditions (figure 4(b)). Once the strain was released and moisture was provided, the resistance recovered close to the initial value, demonstrating that indeed the moisture level plays a critical role in these kinds of experiments. Although humidity control must be considered in benchtop experiments, hydration issue would be non-existent in the envisioned implant applications due to the

water surrounding environment. Another potential issue when characterizing stretchable ion pumps is the movement of the electrodes and the electrolytes, which in some cases generated large artifacts in the measured resistance (figures 4(d), S2 (available online at stacks.iop.org/FPE/6/044004/mmedia)).

Intronic delivery devices can be attractive for applications both on skin, for example in wound healing applications, and as implants within the body. A requirement for both application areas is that the devices must be able to follow the bodily motions with preserved functionality. To demonstrate the practical applicability of our developed stretchable ion pump for these kinds of applications, we used it as a skin patch on the joint of a thumb. The person initially kept the thumb straight (figure 5(a, i)), then bent the thumb slightly (figure 5(a, ii)), and finally bent the thumb to its maximum angle (figure 5(a, iii)) to evaluate the functionality and performance of the device during bodily motions. The subject felt no discomfort during the bending as a result of the device being attached to the thumb. Due to its conformable and stretchable nature, the device sat well on the skin and deformed along with the bending of the thumb (figure 5(a)). The functionality was tested by delivering H^+ from the source to a pH indicator

Figure 4. Characterization of ion pumps under strain. (a) Schematic of the stretching setup for measuring resistance vs strain. (b) Cyclic stretching experiment to different strain levels while measuring the resistance (resistance normalized to initial value) under humid conditions. (c) Extracted relative resistance vs strain from figure (b). For each strain cycle (five at each strain level), the average resistance was calculated for the relaxed ($R_{0,c}$) and strained (R) states, from which the relative resistance $R/R_{0,c}$ was calculated. (d) Long-term resistance changes due to drying of the channel (without elevated humidity). After the cycling, the device was rehydrated at 0% strain.

Figure 5. The ion pump as a stretchable skin patch. (a) The stretchable ion pump is mounted onto the joint of a thumb. (b) The resistance changes with the bending of the thumb, as this induces strain in the device. The labels (i), (ii) and (iii) in (a) correspond to the labeled regions in graph (b).

in the target, thereby confirming continuous delivery during the bending. The ion channel resistance was monitored throughout the experiment and the relative change in resistance is shown in figure 5(b). The resistance was stable for small bending angles (figure 5(b, ii)) but increased by a factor of 2–3 for

maximum bending (iii). Such a limited increase in resistance can easily be compensated by a corresponding increase in driving voltage, thus a stable current and delivery rate can be preserved even under maximum bending. The fast increase in resistance from (ii) to (iii) in figure 5(b) indicates that the change

in channel geometry is the main factor behind the increase, however, changes the channel hydration can also contribute to the slower variations in channel resistance. Altogether, this demonstration indicates the mechanical feasibility of using the developed stretchable ion pump for on skin and implant applications.

4. Conclusions

We have developed an intrinsically stretchable IEM based on A-Hpg, WPU and 2-HEC. With this novel material, we successfully fabricated and tested the first reported fully stretchable ion pump. The ion selectivity of the ion pump was confirmed by three experiments: (a) transport of cations of different diffusion coefficients and comparing with ionic conductivity; (b) changing the anions in the target electrolyte and monitoring the ionic current; and (c) varying the source concentration and monitoring its dependency on the ionic current. The performance of the ion pumps under strain was studied by cyclic stretching experiments while monitoring the ionic current. The ion pumps were found to withstand the cyclic stretching up to 100% strain without any damage to the IEMs, and the ionic resistivity was observed to be stable and reproducible. With a stretchability of up to 100% and a Young's modulus of 41 kPa, the IEM matches the mechanical properties of living tissue well [35]. The presented stretchable IEM can be useful in a wide range of wearable energy harvesting and storage applications, as well as in future soft and stretchable implantable drug delivery devices.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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